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(54) Title: 4-ARYL- AND 4-HETEROARYL -5-OXOPYRAZOLINE DERIVATIVES HAVING PESTICIDAL PROPERTIES

(57) Abstract

The present invention relates to novel, pesticidally effective, 4-aryl- and 4-heteroaryl-5-oxo-pyrazoline derivatives of formula (I), in which R₁, R₂, R₃ and G are as defined in claim 1, to compositions comprising these compounds, to their use as insecticides, acaricides or herbicides, especially in crops of useful plants, and to selective-herbicidal compositions comprising compounds of formula (I) and, as safeners, herbicide-antagonistically effective quantities of either a quinoline derivative of formula (X), in which R₁₅ and X₂ are as defined in the description, or a 1-phenylazole-3-carboxylic acid derivative of formula (XI), in which E, $R_{16},\,R_{17},\,R_{18}$ and R_{19} are as defined in the description.

$$R_{16} \longrightarrow R_{17}$$

$$(XI)$$

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4-Aryl- and 4-heteroaryl -5-oxopyrazoline derivatives having pesticidal properties

The present invention relates to novel, pesticidally active 4-aryl- and 4-heteroaryl-5-oxo-pyrazoline derivatives, to a process for their preparation, to compositions which comprise these compounds as active substances, and to their use as pesticides, especially as insecticides, acaricides or herbicides.

4-Aryl-5-oxo-pyrazolines and 4-aryl- and 4-heteroarylpyrazolidine-3,5-diones having insecticidal, acaricidal and herbicidal action are already known and are described, for example, in EP-A-0 508 126 and WO-92/16510.

Novel 4-aryl- and 4-heteroaryl-5-oxo-pyrazoline derivatives having pesticidal properties have now been found which are distinguished by good activity.

The compounds according to the invention are of the formula

in which

$$R_1$$
 is the group $(R_2)_n$, $(R_5)_m$ or $(R_4)_n$ or $(R_4)_n$

the substituents R_4 independently of one another are halogen, nitro, cyano, C_1 - C_4 alkyl, C_1 - C_4 haloalkyl, C_1 - C_4 haloalkoxy, C_3 - C_6 alkenyloxy, C_1 - C_4 alkoxy- C_2 - C_4 alkoxy, C_3 - C_6 alkynyloxy, C_1 - C_4 alkylcarbonyl, C_1 - C_4 alkylsulfinyl, C_1 - C_4 alkylsulfonyl, amino, C_1 - C_4 alkylsulfinyl, C_1 - C_4 alkylsulfonyl, amino, C_1 - C_4 alkylamino or

 C_1 - C_4 alkylsulfinyl, C_1 - C_4 alkylsulfonyl, amino, C_1 - C_4 alkylamino or di(C_1 - C_4 alkyl)amino;

$$R_5$$
 is the group $-x_1$ or $-x_1$ (R_6) q

n is 0, 1, 2, 3 or 4;

m is 0 or 1, and the sum of m and n is 0, 1, 2, 3 or 4; q is 0, 1, 2 or 3;

 X_1 is oxygen, sulfur, -CH₂- or -N(R₇)-;

the substituents R_6 independently of one another are C_1 - C_4 alkyl, halogen, C_1 - C_4 haloalkyl, C_1 - C_4 alkoxy, C_1 - C_4 haloalkoxy, nitro, cyano, C_1 - C_4 alkoxycarbonyl, amino,

 C_1 - C_4 alkylamino or di(C_1 - C_4 alkyl)amino;

 R_7 is hydrogen, C_1 - C_4 alkyl, formyl or C_1 - C_4 alkylcarbonyl;

R₂ and R₃ independently of one another are hydrogen, C₁-C₈alkyl, C₃-C₈alkenyl,

 C_3 - C_8 alkynyl, C_1 - C_8 alkoxyalkyl, C_1 - C_8 alkylthioalkyl, C_3 - C_8 cycloalkyl or substituted or unsubstituted aryl; or

R₂ and R₃ together form the bivalent radical of a saturated or unsaturated, unsubstituted or substituted, mono-, bi-, tri- or polycyclic system, which system can optionally contain, in positions not adjacent to the nitrogen atoms shown in formula I, one or more ring hetero atoms;

G is the group -CO-A (a) or -SO₂-B (b);

A is C_1 - C_8 alkyl substituted by nitro, cyano, $Si(C_1$ - C_4 alkyl)₃, amino, C_1 - C_4 alkylamino, $di(C_1$ - C_4 alkyl)amino, substituted or unsubstituted benzyloxy or a group $-CH-NR_{08}R_{09}$,

or A is C_3 - C_8 cycloalkyl which can if desired contain as heteroatom oxygen, sulfur or nitrogen and is substituted by C_1 - C_4 alkyl, C_1 - C_4 alkoxy, C_1 - C_4 alkylthio,

 C_1 - C_4 alkylsulfinyl, C_1 - C_4 alkylsulfonyl or substituted or unsubstituted phenyl, or A is C_3 - C_8 cycloalkyloxy which can if desired contain as heteroatom oxygen, sulfur or nitrogen and is substituted by C_1 - C_4 alkyl, or A is adamantyl, naphthyl, naphthyl substituted by halogen, C_1 - C_4 alkyl, C_1 - C_4 haloalkyl or C_1 - C_4 alkoxy, or A is a group

 $-(CR_8R_9)_p-(Y)_o-CO-(Z)_r-R_{10}$, $-(CR_8R_9)_p-R_{11}$ or $-N(R_{10})R_{13}$;

 R_{08} , R_{09} and R_{010} independently of one another are hydrogen or C_1 - C_6 alkyl;

R₈ and R₉ independently of one another are hydrogen, C₁-C₆alkyl or phenyl;

 R_{10} is hydrogen, C_1 - C_{10} alkyl, C_1 - C_{10} alkyl substituted by halogen, C_1 - C_4 alkoxy,

 C_1 - C_4 haloalkoxy, C_1 - C_4 alkylthio, C_3 - C_{12} cycloalkyl, cyano, aryl or aryloxy,

C₃-C₈cycloalkyl, C₃-C₈cycloalkyl substituted by halogen or C₁-C₄alkyl, C₃-C₁₀alkenyl,

C₃-C₁₀alkynyl, C₃-C₁₀haloalkenyl, C₃-C₁₀haloalkynyl, aryl, aryl substituted by halogen,

 C_1 - C_4 alkyl, C_1 - C_4 haloalkyl, C_1 - C_6 alkoxy, C_1 - C_4 haloalkoxy, C_1 - C_4 alkylthio, nitro, cyano, phenoxy or halo-substituted phenoxy;

o and r independently of one another are 0 or 1;

Y and Z independently of one another are oxygen or NR_{12} ; p is 1 to 10:

 R_{11} is C_3 - C_8 cycloalkyl, C_3 - C_8 cycloalkyl substituted by halogen or C_1 - C_4 alkyl, aryloxy, aryloxy substituted by halogen, C_1 - C_4 alkyl, C_1 - C_4 alkoxy,

 C_1 - C_4 haloalkoxy, C_1 - C_4 alkylthio, nitro, cyano, phenoxy or halo-substituted phenoxy, benzyloxy substituted by halogen, C_1 - C_4 alkyl, C_1 - C_4 haloalkyl, C_1 - C_4 alkoxy, C_1 - C_4 haloalkoxy, C_1 - C_4 alkylthio, nitro, cyano, phenoxy or halo-substituted phenoxy, norbornyl or adamantyl, where aryloxy with the exception of phenoxy can in each case be

any desired aryloxy group;

 R_{12} is hydrogen, C_1 - C_4 alkyl, C_3 - C_8 cycloalkyl, C_3 - C_{10} alkenyl or C_3 - C_{10} alkynyl; R_{13} is C_3 - C_8 cycloalkyl, C_3 - C_8 cycloalkyl substituted by halogen or C_1 - C_4 alkyl, or cyano- C_1 - C_6 alkyl;

B is C₁-C₁₀alkyl substituted by C₁-C₄alkoxy, C₁-C₆alkoxy-C₁-C₄alkoxy,

C₁-C₄haloalkoxy, C₁-C₄alkylthio, C₁-C₄haloalkylthio, C₁-C₄alkylsulfinyl,

 C_1 - C_4 alkylsulfonyl, C_1 - C_4 alkylsulfonyloxy, C_1 - C_4 alkylcarbonyl, C_1 - C_4 alkylcarbonyloxy, di(C_1 - C_4 alkyl)amino, C_3 - C_8 cycloalkyl, aryl, aryloxy, arylthio, arylsulfonyl, arylsulfonyloxy, arylcarbonyl or pyridyl, it being possible for

the aryl and pyridyl groups if desired to be substituted by halogen, C_1 - C_4 alkyl,

 C_1 - C_4 haloalkyl, C_1 - C_4 alkoxy, C_1 - C_4 haloalkoxy, C_1 - C_4 alkylthio, nitro, cyano, phenoxy, halo-substituted phenoxy, phenylthio or halo-substituted phenylthio, or B is

 C_3 - C_{10} alkenyl, C_3 - C_{10} haloalkenyl, C_3 - C_{10} haloalkynyl, C_3 - C_8 cycloalkyl, C_3 - C_8 cycloalkyl substituted by halogen or C_1 - C_4 alkyl, benzyl, benzyl substituted by

halogen, C₁-C₄alkyl, C₁-C₄haloalkyl or C₁-C₄alkoxy, naphthyl, naphthyl substituted by halogen or nitro, or B is a group -N(R₂₁)R₂₂:

R₂₁ is C₁-C₁₀alkyl substituted by C₁-C₄alkoxy, C₁-C₆alkoxy-C₁-C₄alkoxy,

 C_1 - C_4 haloalkoxy, C_1 - C_4 alkylthio, C_1 - C_4 haloalkylthio, C_1 - C_4 alkylsulfinyl,

 C_1 - C_4 alkylsulfonyl, C_1 - C_4 alkylsulfonyloxy, C_1 - C_4 alkylcarbonyl, C_1 - C_4 alkoxycarbonyl,

 C_1 - C_4 alkylcarbonyloxy, di(C_1 - C_4 alkyl)amino, C_3 - C_8 cycloalkyl, aryl, aryloxy, arylthio, arylsulfonyl, arylsulfonyloxy, arylcarbonyl or pyridyl, it being possible for

the aryl and pyridyl groups if desired to be substituted by halogen, C₁-C₄alkyl,

 C_1 - C_4 haloalkyl, C_1 - C_4 alkoxy, C_1 - C_4 haloalkoxy, C_1 - C_4 alkylthio, nitro, cyano, phenoxy, halo-substituted phenoxy, phenylthio or halo-substituted phenylthio, or R_{21} is

C₃-C₂₀alkenyl, C₃-C₂₀haloalkenyl, C₃-C₂₀alkynyl, C₃-C₂₀haloalkynyl, C₃-C₈cycloalkyl,

 C_3 - C_8 cycloalkyl substituted by halogen or C_1 - C_4 alkyl, benzyl or aryl, it being possible for the benzyl and aryl groups if desired to be substituted by halogen, C_1 - C_4 alkyl,

 C_1 - C_4 haloalkyl, C_1 - C_4 alkoxy, C_1 - C_4 haloalkoxy, C_1 - C_4 alkylthio, nitro, cyano, benzoyl, halo-substituted benzoyl, phenoxy or halo-substituted phenoxy;

R₂₂ is as defined for R₂₁ or else alternatively is C₁-C₁₀alkyl; or

 R_{21} and R_{22} , together with the nitrogen atom to which they are attached, form an unsubstituted or mono- to trisubstituted, nonaromatic, mono- or bicyclic heterocyclic ring in which the substituents can be C_1 - C_4 alkyl, C_1 - C_4 alkyl substituted by halogen,

C₁-C₄alkoxy, phenyl, benzodioxoyl or trifluoromethylphenyl, C₁-C₄alkoxy,

C₁-C₄alkoxycarbonyl, C₁-C₄alkylcarbonyl, phenyl, phenyl substituted by halogen,

C₁-C₄alkyl, C₁-C₄alkoxy, nitro or trifluoromethyl, pyridyl, pyrimidinyl or formyl, and salts and diastereomers of the compounds of the formula I, with the proviso that, if R₂₁

and R_{22} are alkyl, at least one of these radicals is a substituted alkyl.

In the above definitions, halogen (halo) refers to iodine and preferably to fluorine, chlorine and bromine.

Unless specifically mentioned, the alkyl, alkenyl and alkynyl groups preferably have 1 or, respectively, 2 to 8 carbon atoms and can be straight-chain or branched, specifications which also apply to the alkyl, alkenyl and alkynyl moieties of alkylsilanes, haloalkyl, alkoxy, haloalkoxy, alkoxyalkyl, alkoxyalkoxy, alkenyloxy, alkynyloxy, alkylcarbonyl, alkoxycarbonyl, alkylthio, alkenylthio, alkynylthio, alkylthioalkyl, alkylsulfinyl, alkylsulfonyl, alkylamino and dialkylamino groups.

Examples of the alkyl groups occurring in the definitions of the substituents are methyl, ethyl, n-propyl, isopropyl, n-butyl, sec-butyl, isobutyl and tert-butyl, and the isomeric pentyls, hexyls, heptyls and octyls. The alkyl groups occurring as or in the substituents preferably have 1-8 carbon atoms.

Alkenyl refers for example to allyl, methallyl, 1-methylvinyl or but-2-en-1-yl. Preferred alkenyl radicals are those having a chain length of 2 to 8 carbon atoms. The alkenyl radicals are preferably attached to a heteroatom via a saturated carbon atom.

Examples of alkynyl are propargyl, but-2-yn-1-yl, 2-methylbutyn-2-yl, but-3-yn-2-yl and pent-4-yn-1-yl. Preferred alkynyl radicals are those having a chain length of 2 to 8 carbon atoms. The alkynyl radicals are preferably attached to a heteroatom via a saturated carbon atom.

Haloalkyl radicals preferably have a chain length of 1 to 8 carbon atoms. Haloalkyl is for example fluoromethyl, difluoromethyl, trifluoromethyl, chloromethyl, dichloromethyl, trichloromethyl, 2,2,2-trifluoroethyl, 2-fluoroethyl, 2-chloroethyl, pentafluoroethyl, 1,1-difluoro-2,2,2-trichloroethyl, 2,2,3,3-tetrafluoroethyl and 2,2,2-trichloroethyl; preference is given to trichloromethyl, difluorochloromethyl, difluoromethyl, trifluoromethyl and dichlorofluoromethyl.

Suitable haloalkenyl groups are alkenyl groups substituted one or more times by halogen, in which context halogen is bromine, iodine and, in particular, fluorine and chlorine, examples being 2,2-difluoro-1-methylvinyl, 3-fluoropropenyl, 3-chloropropenyl, 3-bromopropenyl, 2,3,3-trifluoropropenyl, 2,3,3-trifluoropropenyl,

 C_3 - C_{20} alkenyl radicals substituted 1, 2 or 3 times by halogen, preference is given to those having a chain length of 3 to 5 carbon atoms.

Suitable haloalkynyl groups are, for example, alkynyl groups which are mono- or polysubstituted by halogen, where halogen is bromine, iodine and, in particular, fluorine and chlorine, examples being 3-fluoropropynyl, 3-chloropropynyl, 3-bromopropynyl, 3,3,3-trifluoropropynyl and 4,4,4-trifluorobut-2-yn-1-yl. Among the alkynyl radicals substituted one or more times by halogen, preference is given to those having a chain length of 3 to 5 carbon atoms.

Alkoxy radicals preferably have a chain length of 1 to 6 carbon atoms. Alkoxy is for example methoxy, ethoxy, propoxy, i-propoxy, n-butoxy, isobutoxy, sec-butoxy and tert-butoxy and the isomers of pentyloxy and hexyloxy; methoxy and ethoxy are preferred.

Alkylcarbonyl is especially acetyl and propionyl.

Alkoxycarbonyl is methoxycarbonyl, ethoxycarbonyl, propoxycarbonyl, isopropoxycarbonyl, n-butoxycarbonyl, isobutoxycarbonyl, sec-butoxycarbonyl and tert-butoxycarbonyl; methoxycarbonyl and ethoxycarbonyl are preferred.

Haloalkoxy radicals preferably have a chain length of 1 to 8 carbon atoms. Haloalkoxy is for example fluoromethoxy, difluoromethoxy, trifluoromethoxy, 2,2,2-trifluoroethoxy, 1,1,2,2-tetrafluoroethoxy, 2-fluoroethoxy, 2-chloroethoxy, 2,2-difluoromethoxy and 2,2,2-trichloroethoxy; difluoromethoxy, 2-chloroethoxy and trifluoromethoxy are preferred.

Alkylthio radicals preferably have a chain length of 1 to 8 carbon atoms. Alkylthio is for example methylthio, ethylthio, propylthio, isopropylthio, n-butylthio, isobutylthio, sec-butylthio or tert-butylthio; methylthio and ethylthio are preferred.

Alkylsulfinyl is methylsulfinyl, ethylsulfinyl, propylsulfinyl, isopropylsulfinyl, n-butylsulfinyl, isobutylsulfinyl, sec-butylsulfinyl, tert-butylsulfinyl; methylsulfinyl and ethylsulfinyl are preferred.

Alkylsulfonyl is methylsulfonyl, ethylsulfonyl, propylsulfonyl, isopropylsulfonyl, n-butylsulfonyl, isobutylsulfonyl, sec-butylsulfonyl, tert-butylsulfonyl; methylsulfonyl and ethylsulfonyl are preferred.

Alkoxyalkoxy radicals preferably have a chain length of 1 to 8 carbon atoms. Examples of alkoxyalkoxy are methoxymethoxy, methoxyethoxy, methoxypropoxy, ethoxymethoxy, ethoxyethoxy, propoxymethoxy and butoxybutoxy.

Alkylamino is for example methylamino, ethylamino, n-propylamino, isopropylamino and the isomeric butylamines.

Dialkylamino is for example dimethylamino, methylethylamino, diethylamino, n-propylmethylamino, dibutylamino and diisopropylamino. Preference is given to alkylamino radicals having a chain length of 1 to 4 carbon atoms.

Alkoxyalkyl radicals preferably have 1 to 8 carbon atoms. Alkoxyalkyl is for example methoxymethyl, methoxyethyl, ethoxymethyl, ethoxyethyl, n-propoxymethyl, n-propoxymethyl, isopropoxymethyl and isopropoxyethyl.

Alkylthioalkyl radicals preferably have 1 to 8 carbon atoms. Alkylthioalkyl is for example methylthiomethyl, methylthioethyl, ethylthiomethyl, ethylthioethyl, n-propylthiomethyl, n-propylthioethyl, isopropylthioethyl, butylthiomethyl, butylthioethyl or butylthiobutyl.

The cycloalkyl radicals preferably have 3 to 8 ring carbon atoms, for example cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl, cycloheptyl and cyclooctyl. These cycloalkyl radicals can if desired be interrupted by heteroatoms such as, for example, oxygen atoms and/or sulfur atoms and/or nitrogen atoms. Similar comments apply to the cycloalkyloxy radicals.

Phenyl, on its own or as part of a substituent such as phenoxy, benzyl, benzyloxy, benzoyl, phenylthio, phenylalkyl or phenoxyalkyl, can in general be unsubstituted or substituted. In the latter case the substituents can be in an ortho, meta and/or para position. Preferred substituent positions are the ortho and para positions relative to the ring linking site. Preferred substituents are halogen, nitro, cyano, C_1 - C_4 alkoxy, C_1 - C_4 haloalkythio, C_1 - C_4 - C_4 haloalkythio, C_1 - C_4 -

Aryl in the definition of the radicals R_2 , R_3 and R_{10} is α - or β -naphthyl, especially phenyl, which aromatic ring systems can carry one or more identical or different substituents, examples being halogen, nitro, cyano, C_1 - C_4 alkoxy, C_1 - C_4 haloalkoxy, C_1 - C_4 alkylthio, C_1 - C_4 haloalkylthio, C_1 - C_4 alkyl and C_1 - C_4 haloalkyl. Corresponding comments apply to

aryloxy, arylthio, arylsulfonyl, arylsulfonyl, arylsulfonyloxy and arylcarbonyl in the definition of the radicals R_{10} , R_{11} , R_{21} , R_{22} and B.

Examples of the unsubstituted or mono- to trisubstituted, nonaromatic, mono- or bicyclic heterocyclic rings which the substituents R_{21} and R_{22} , together with the nitrogen atom to which they are attached, are able to form are, for example, 1-pyrrolidinyl, 1-piperidyl, 2,6-dimethyl-1-piperidyl, 4-morpholinyl, 4-methylpiperazin-1-yl, 4-phenylpiperazin-1-yl, 4-benzylpiperazin-1-yl, 4-(3,4-dichlorophenyl)piperazin-1-yl, 4-thiomorpholinyl, 2,6-dimethyl-4-morpholinyl, 2,6-dimethyl-thio-4-morpholinyl, 1,2,3,4-tetrahydroquinolin-1-yl or indolin-1-yl.

Examples of saturated or unsaturated and unsubstituted or substituted mono-, bi-, tri- or polycyclic systems which the substituents R_2 and R_3 , together with the two nitrogen atoms of the pyrazoline ring to which they are attached, are able to form are:

These ring systems can be substituted by halogen, C_1 - C_{16} alkyl, C_1 - C_{16} haloalkyl, C_2 - C_{16} alkenyl, C_1 - C_{16} alkoxy- C_2 - C_6 alkyl, C_1 - C_6 polyalkoxy- C_2 - C_6 alkyl or by phenyl or benzyl each of which is unsubstituted or substituted by halogen, nitro, C_1 - C_4 alkyl, C_1 - C_3 alkoxy or C_1 - C_3 haloalkyl; and R_{14} is halogen or unsubstituted or halo-substituted C_1 - or C_2 alkoxy.

Corresponding meanings can also be assigned to the substituents in composite definitions, for example haloalkoxy, haloalkylthio, alkoxyalkoxy, alkoxycarbonyl, alkylcarbonyl, alkylsulfinyl, alkylsulfonyl, alkenyloxy, alkynyloxy, alkenylthio, alkynylthio and cycloalkylthio.

The invention likewise comprises the salts which the compounds of the formula I can form with acids. Suitable acids for the formation of the acid addition salts are both organic and inorganic acids. Examples of such acids are inter alia hydrochloric acid, hydrobromic acid, nitric acid, various phosphorus acids, sulfuric acid, acetic acid, propionic acid, butyric acid, valeric acid, oxalic acid, malonic acid, fumaric acid, organic sulfonic acids, lactic acid, tartaric acid, citric acid and salicylic acid.

The salts of the compounds of the formula I containing acidic hydrogen, as for example in the definition of the substituent A as C_1 - C_8 alkyl substituted by a group $-\frac{CH-NR_{08}R_{09}}{COOR_{010}}$, in

which R₀₁₀ is hydrogen, can also be alkali metal salts, for example sodium salts and potassium salts; alkaline earth metal salts, for example calcium salts and magnesium salts; ammonium salts, i.e. unsubstituted ammonium salts and mono- or polysubstituted ammonium salts, and salts with other organic nitrogen bases.

Correspondingly, suitable salt formers are alkali metal hydroxides and alkaline earth metal hydroxides, especially the hydroxides of lithium, sodium, potassium, magnesium or calcium, with particular importance being attached to those of sodium or potassium.

Examples of amines suitable for forming ammonium salts include not only ammonia but also primary, secondary and tertiary C₁-C₁₈alkylamines, C₁-C₄hydroxyalkylamines and C₂-C₄alkoxyalkylamines, for example methylamine, ethylamine, n-propylamine, isopropylamine, the four isomeric butylamines, n-amylamine, isoamylamine, hexylamine, heptylamine, octylamine, nonylamine, decylamine, pentadecylamine, hexadecylamine, heptadecylamine, octadecylamine, methylethylamine, methylisopropylamine, methylhexylamine, methylnonylamine, methylpentadecylamine, methyloctadecylamine, ethylbutylamine, ethylheptylamine, ethyloctylamine, hexylheptylamine, hexyloctylamine, dimethylamine, diethylamine, di-n-propylamine, diisopropylamine, di-n-butylamine. di-n-amylamine, diisoamylamine, dihexylamine, diheptylamine, dioctylamine, ethanolamine, n-propanolamine, isopropanolamine, N,N-diethanolamine, N-ethylpropanolamine, N-butylethanolamine, allylamine, n-butenyl-2-amine, n-pentenyl-2-amine, 2,3-dimethylbutenyl-2-amine, dibutenyl-2-amine, n-hexenyl-2-amine, propylenediamine, trimethylamine, triethylamine, tri-n-propylamine, tri-isopropylamine, tri-n-butylamine, triisobutylamine, tri-sec-butylamine, tri-n-amylamine, methoxyethylamine and ethoxyethylamine; heterocyclic amines such as, for example, pyridine, quinoline, isoquinoline, morpholine, N-methylmorpholine, thiomorpholine,

piperidine, pyrrolidine, indoline, quinuclidine and azepine; primary arylamines such as, for example, anilines, methoxyanilines, ethoxyanilines, o-, m- and p-toluidines, phenylenediamines, benzidines, naphthylamines and o-, m- and p-chloroanilines; but especially triethylamine, isopropylamine and diisopropylamine.

In addition, it is possible if desired to form corresponding inner salts, for example in the case of the radical A as C_1 - C_8 alkyl substituted by a group $-CH-NR_{08}R_{09}$, in which R_{08} ,

R₀₉ and R₀₁₀ are hydrogen.

Furthermore, it is possible for certain substituents R₁, R₄ to R₁₃, and R₂ and R₃ alone or in combination with one another or in combination with the parent structure to which they are attached to exhibit chirality. The invention comprises both the racemate and the enriched and optically pure forms of the respective stereoisomers.

In the processes described in this application, the asymmetrically substituted compounds of the formula I are generally - unless chiral precursors are used - obtained as racemates. The stereoisomers can then be separated on the basis of physicochemical properties by known methods, for instance fractional crystallization after the formation of salts with optically pure bases, acids or metal complexes, or else by chromatographic methods such as, for example, high-performance liquid chromatography (HPLC) on acetylcellulose. In the present invention, references to the active substances of the formula I include both the pure optical isomers and the racemates and/or diastereomers. Where no specific reference to the individual optical isomers is made, the formula given should be taken to refer to those racemic mixtures which are produced in the preparation process indicated. Where an aliphatic C=C double bond is present, geometric isomerization can also occur.

The formula I is intended to embrace all of these possible isomers and mixtures thereof.

With respect to the insecticidal and acaricidal actions, emphasis should be put on the following compounds of the formula I:

Preferred compounds of the formula I are those in which R₁ is the group

 $(R_4)_n$. Of these compounds, particular preference is given to those in which $(R_5)_m$

R₄ is fluorine, chlorine or C₁-C₄alkyl; n is 1, 2 or 3; and m is 0.

Of these compounds, very particular preference is given to those in which R_4 is fluorine, chlorine or methyl; and n is 2 or 3.

Preference is likewise given to compounds in which R_2 and R_3 are C_1 - C_6 alkyl; or R_2 and R_3 together form a substituted or unsubstituted alkylene chain -(CH_2)₅-, -(CH_2)₄- or -(CH_2)₃-.

Compounds which are likewise preferred are those in which G is the group -CO-A (a). Among these compounds, particular preference is given to those in which A is a group - $(CR_8R_9)_p$ - $(Y)_o$ -CO- $(Z)_r$ - R_{10} , - $(CR_8R_9)_p$ - R_{11} or - $N(R_{10})R_{13}$, or substituted or unsubstituted naphthyl.

Among these compounds, very particular preference is given to those in which R_8 and R_9 independently of one another are hydrogen or methyl; and p is 1 or 2.

Of these compounds, very special importance attaches to those in which R_8 and R_9 are methyl.

Likewise particularly preferred are compounds in which R_{10} is C_1 - C_8 -alkyl, preferably C_1 - C_4 -alkyl, C_3 - C_6 cycloalkyl, phenyl, phenyl substituted by halogen, or is naphthyl; R_{11} is C_3 - C_8 cycloalkyl, benzyloxy, benzyloxy substituted by halogen, C_1 - C_4 alkyl, C_1 - C_4 haloalkyl or phenoxy, or is naphthyloxy; and R_{13} is C_3 - C_7 cycloalkyl or cyano- C_1 - C_6 alkyl.

Other particularly preferred compounds are those in which Y and Z are NR_{12} . Among these compounds, particular preference is given to those in which R_{12} is hydrogen or C_1 - C_4 alkyl.

Particular preference is similarly given to compounds in which R_8 and R_9 independently of one another are hydrogen or methyl; p is 1 or 2; R_{10} is C_1 - C_4 alkyl, C_3 - C_6 cycloalkyl, phenyl, phenyl substituted by halogen, or is naphthyl; R_{11} is C_3 - C_6 cycloalkyl, benzyloxy, benzyloxy substituted by halogen, C_1 - C_4 alkyl, C_1 - C_4 haloalkyl or phenoxy, or is naphthyloxy; Y and Z are NR_{12} ; R_{12} is hydrogen or C_1 - C_4 alkyl; and R_{13} is C_3 - C_8 cycloalkyl or cyano- C_1 - C_6 alkyl.

Preferred compounds are those in which R_1 is the group $(R_4)_n$; R_4 is fluorine,

chlorine or C_1 - C_4 alkyl; n is 1, 2 or 3; m is 0; R_2 and R_3 are C_1 - C_6 alkyl; or R_2 and R_3 together form a substituted or unsubstituted alkylene chain - $(CH_2)_5$ -, - $(CH_2)_4$ - or - $(CH_2)_3$ -; G is the group -CO-A (a); A is a group - $(CR_8R_9)_p$ - $(Y)_o$ -CO- $(Z)_r$ - R_{10} , - $(CR_8R_9)_p$ - R_{11} or - $N(R_{10})R_{13}$, or is unsubstituted or substituted naphthyl; R_8 and R_9 independently of one another are hydrogen or methyl; p is 1 or 2; R_{10} is C_1 - C_4 alkyl, C_3 - C_6 cycloalkyl, phenyl, phenyl substituted by halogen, or is naphthyl; R_{11} is C_3 - C_8 cycloalkyl, benzyloxy, benzyloxy; and R_{13} is C_3 - C_8 cycloalkyl or cyano- C_1 - C_6 alkyl.

With regard to the herbicidal properties, preferred compounds of the formula I are those following in which R_1 is the group $(R_4)_n$; and m is 0. Among these

compounds, particular preference is given to those following in which R_4 is C_1 - or C_2 alkyl; and n is 1, 2 or 3.

Among these compounds of the formula I, very particular preference is given to those in which R_4 is C_1 - or C_2 alkyl in positions 2, 4 and 6 relative to the phenyl linkage site; and n is 3.

Other preferred compounds are those in which R_1 is the group $(R_4)_n$; m is 0;

 R_4 is C_1 - or C_2 alkyl; n is 1, 2 or 3; R_2 and R_3 are C_1 - C_6 alkyl; or R_2 and R_3 together form a substituted or unsubstituted alkylene chain -(CH_2)₅-, -(CH_2)₄- or -(CH_2)₃-.

Likewise preferred are compounds in which G is a group -CO-A (a); and A is C_1 - C_8 alkyl substituted by nitro, cyano, $Si(C_1$ - C_4 alkyl)₃, amino, C_1 - C_4 alkylamino, $di(C_1$ - C_4 alkyl)amino, substituted or unsubstituted benzyloxy or a group $-CH-NR_{08}R_{09}$, $COOR_{010}$

or A is C_3 - C_8 cycloalkyl, which can if desired contain as heteroatom oxygen, sulfur or nitrogen and is substituted by C_1 - C_4 alkyl, C_1 - C_4 alkoxy, C_1 - C_4 alkylsulfinyl, C_1 - C_4 alkylsulfinyl or substituted or unsubstituted phenyl, or A is C_3 - C_8 cycloalkyloxy which can if desired contain as heteroatom oxygen, sulfur or nitrogen and is unsubstituted or substituted by C_1 - C_4 alkyl, or A is a group

 $-(CR_8R_9)_p-(Y)_o-CO-(Z)_r-R_{10} \text{ or } -(CR_8R_9)_p-R_{11}.$

Other preferred compounds are those in which R_1 is the group (R_2) ; R_3

or C₂alkyl; n is 1, 2 or 3; m is 0; R₂ and R₃ are C₁-C₆alkyl; or R₂ and R₃ together form a substituted or unsubstituted alkylene chain -(CH₂)₅-, -(CH₂)₄- or -(CH₂)₃-; G is the group -CO-A-(a); and A is C₁-C₈alkyl substituted by nitro, cyano, Si(C₁-C₄alkyl)₃, amino, C₁-C₄alkylamino, di(C₁-C₄alkyl)amino, substituted or unsubstituted benzyloxy or a group -CH-NR₀₈R₀₉, or A is C₃-C₈cycloalkyl which can if desired contain as heteroatom COOR₀₁₀

oxygen, sulfur or nitrogen and is substituted by C_1 - C_4 alkyl, C_1 - C_4 alkoxy, C_1 - C_4 alkylsulfinyl, C_1 - C_4 alkylsulfonyl or substituted or unsubstituted phenyl, or A is C_3 - C_8 cycloalkyloxy which can if desired contain as heteroatom oxygen, sulfur or nitrogen and is unsubstituted or substituted by C_1 - C_4 alkyl, or A is a group - $(CR_8R_9)_p$ - $(Y)_o$ -CO- $(Z)_r$ - R_{10} or - $(CR_8R_9)_p$ - R_{11} .

Especially preferred is the compound of the formula I, in which R_1 is 2,4,6-trimethylphenyl, R_2 and R_3 together are -(CH₂)₄- and G is -C(=O)-N(CH₃)CH₂CH₂CN.

Also especially preferred is the compound of the formula I, in which R_1 is 2,4,6-trimethylphenyl, R_2 and R_3 -N together are -CH₂CH₂CH(CH₃)CH₂-N and G is -C(=O)-N(CH₃)CH₂CN.

Also especially preferred is the compound of the formula I, in which R_1 is 2,6-dichlorophenyl, R_2 and R_3 together are -(CH₂)₄- and G is -C(=O)-N(CH₃)CH₂CN.

The process according to the invention for the preparation of compounds of the formula I is carried out in analogy to known processes and comprises

a) for the preparation of the 4-aryl- and 4-heteroaryl-5-oxopyrazoline derivatives of the formula

$$\begin{array}{c}
R_3 \\
N \\
R_2
\end{array}$$

$$\begin{array}{c}
N \\
R_1
\end{array}$$
(I),

in which R₁, R₂ and R₃ are as defined under formula I, G is the group -CO-A (a) and A is

as defined under formula I with the exception of the group -N(R₁₀)R₁₃, reacting a compound of the formula

$$R_3$$
 N
 R_2
 N
 R_1
 R_1
 R_1
 R_1
 R_1

in which R₁, R₂ and R₃ are as defined, with a compound of the formula

$$\begin{array}{ccc}
O \\
A - C - E_1
\end{array} (III)$$

in which A is as defined and E1 is a leaving group, preferably halogen and especially chlorine;

b) for the preparation of the 4-aryl- and 4-heteroaryl-5-oxopyrazoline derivatives of the formula

$$R_3$$
 R_2
 R_1
 R_1
 R_1
 R_1
 R_1

in which R₁, R₂ and R₃ are as defined under formula I, G is the group -CO-A (a), and A is -N(R_{10}) R_{13} , where R_{10} and R_{13} are as defined under formula I with the exception of R_{10} as hydrogen, reacting a compound of the formula

$$R_3$$
 R_2
 N
 R_1
 R_1
 R_1
 R_2
 R_1
 R_1

in which
$$R_1$$
, R_2 and R_3 are as defined, with a compound of the formula
$$E_1 - \overset{O}{C} - N \overset{R_{10}}{\underset{R_{13}}{\vee}}$$
 (IV),

in which R_{10} and R_{13} are as defined and E_1 is a leaving group, preferably halogen and especially chlorine; or

c) for the preparation of the 4-aryl- and 4-heteroaryl-5-oxopyrazoline derivatives of the formula

$$R_3$$
 R_2
 N
 R_1
 R_1
 R_1
 R_1

in which R_1 , R_2 and R_3 are as defined under formula I, G is the group -CO-A (a), and A is -NHR₁₃, where R_{13} is as defined under formula I, reacting a compound of the formula

$$R_3$$
 N R_1 R_2 R_1 (II),

in which R₁, R₂ and R₃ are as defined, with an isocyanate of the formula

$$R_{13}-N=C=O (V)$$

in which R₁₃ is as defined.

The process according to the invention for the preparation of compounds of the formula

in which R_1 , R_2 and R_3 are as defined under formula I, G is the group -SO₂-B (b), and B is as defined under formula I, is carried out in analogy to known processes as described, for example, in EP-A-0 508 126, and comprises reacting a compound of the formula

$$\begin{array}{c}
R_3 \\
N \\
R_2
\end{array}$$
OH
$$R_1$$
(II),

in which R_1 , R_2 and R_3 are as defined under formula I, with a compound of the formula B-SO₂-E₁ (VI),

in which B is as defined and E₁ is a leaving group such as halogen, especially chlorine, if desired in a solvent and in the presence or absence of a base.

The reactions according to process variants (a), (b) and (c) can be carried out in analogy to known processes, advantageously in a reaction-inert, hydroxyl-free solvent in the presence of an organic base such as, for example, pyridine, 4-dimethylaminopyridine, lutidine, collidine, trialkylamines, N,N-dialkylaniline, or bicyclic nonnucleophilic bases such as,

for example, 1,4-diazabicyclo[2.2.2]octane (DABCO), 1,5-diazabicyclo[4.3.0]non-5-ene (DBN) or 1,8-diazabicyclo[5.4.0]undec-7-ene (DBU). The reaction is generally carried out at temperatures of from 0 to 120°C, preferably from 20 to 90°C, advantageously in the presence of a reaction-inert solvent or solvent mixture. Examples of those suitable for this purpose are aliphatic and aromatic hydrocarbons such as benzene, toluene, petroleum ether or hexane; halogenated hydrocarbons such as chlorobenzene, methylene chloride, ethylene chloride, chloroform, carbon tetrachloride and tetrachloroethylene; ethers and ether-like compounds, such as dialkyl ethers, for example diethyl ether, diisopropyl ether and tert-butyl methyl ether, and also anisole, dioxane and tetrahydrofuran; nitriles such as acetonitrile and propionitrile; esters such as ethyl acetate, propyl acetate and butyl acetate; ketones such as acetone, diethyl ketone and methyl ethyl ketone; and mixtures of such solvents.

The reaction of the compound of the formula II with the reactive sulfonic acid derivative of the formula VI takes place advantageously in equimolar quantities at temperatures of from 0 to 150°C, preferably from 20 to 70°C. Suitable solvents are the customary reaction-inert, polar, organic solvents, such as ethers, amides, nitriles, alcohols, sulfones or sulfoxides, especially dimethyl sulfoxide, tetrahydrofuran or N,N-dimethylformamide. Suitable binding agents for the acid formed are the known inorganic and organic bases such as sodium hydroxide, sodium carbonate, potassium carbonate or pyridines. The reaction can take place under elevated pressure or, in particular, at atmospheric pressure. If desired it is also possible to operate under phase transfer conditions, in which case from 0.3 to 1.5 mol of reactive sulfonic acid derivative of the formula VI are employed per mole of the compound of the formula II at from 0 to 150°C, preferably from 20 to 70°C. Phase transfer catalysts which can be used are the customary quaternary ammonium salts, such as tetraoctylammonium bromide or benzyltriethylammonium chloride. In this case suitable organic solvents are all inert apolar solvents, such as benzene or toluene.

The compounds of the formula II, in which R_1 , R_2 and R_3 are as defined under formula I, are known from EP-A-0 508 126 and WO 92/16510 and can be prepared by the processes disclosed.

The compounds of the formulae III, IV, V and VI required for the preparation processes are either commercial products or can be prepared in analogy to known standard processes.

The end products of the formula I can be isolated in a customary manner by concentration

and/or evaporation of the solvent and purified by recrystallization or trituration of the solid residue in solvents in which they are not readily soluble, such as ethers, alkanes, aromatic hydrocarbons or chlorinated hydrocarbons, or by means of chromatography over a silica gel column.

Salts of compounds of the formula I can be prepared in a manner known per se. Thus, for example, acid addition salts of compounds of the formula I are obtained by treatment with a suitable acid or a suitable ion exchanger reagent, and salts with bases by treatment with a suitable base or a suitable ion exchanger reagent.

Salts of compounds of the formula I can be converted into the free compounds of the formula I in a customary manner; acid addition salts, for example, by treatment with a suitable basic agent or a suitable ion exchanger reagent, and salts with bases, for example, by treatment with a suitable acid or a suitable ion exchanger reagent.

Salts of compounds of the formula I can be converted into other salts of compounds of the formula I in a manner known per se; acid addition salts, for example, can be converted into other acid addition salts, for example, by treating a salt of an inorganic acid, such as a hydrochloride, with an appropriate metal salt, such as a sodium, barium or silver salt, of an acid, for example with silver acetate, in a suitable solvent in which the inorganic salt which forms, for example silver chloride, is insoluble and therefore separates out of the reaction mixture.

Depending on the procedure and reaction conditions, the compounds of the formula I with salt-forming properties can be obtained in free form or in the form of salts.

Unless specific synthesis is carried out for the isolation of pure isomers, the product can be obtained as a mixture of two or more isomers. The isomers can be separated by methods known per se.

For the compositions comprising the compounds of the formula I, the preferences which apply are the same as those for the compounds of the formula I.

The present invention also relates to a selective-herbicidal composition for controlling grasses and weeds in crops of useful plants, especially in crops of maize and cereal, which composition comprises a herbicide and a safener (counteracting agent, antidote) and which protects the useful plants but not the weeds against the phytotoxic action of the herbicide,

and to the use of this composition for weed control in crops of useful plants.

When herbicides are used it is possible, for example, depending on the rate and mode of application of the herbicide, on the crop plant, on the nature of the soil and on the climatic conditions, such as period of exposure to light, temperature and amounts of precipitation, for the crop plants as well to be damaged to a considerable extent.

In order to counter this and similar problems, a variety of substances have already been proposed as safeners, which are capable of antagonizing the damaging effect of the herbicide on the crop plant, in other words of protecting the crop plant from said effect, while causing virtually no impairment of the herbicidal action on the weeds to be controlled. In this context it has been found that the proposed safeners often have a very specific action with respect to the crop plants and with respect to the herbicide and, in some cases, in dependence on the mode of application; in other words, a certain safener is often suitable only for one specific crop plant and one specific class of herbicide or a specific herbicide. Thus, for example, EP-A-0 094 349 discloses quinoline derivatives which protect crop plants against the phytotoxic effect of herbicides of particular classes, for example chloroacetanilides, phenoxypropionates, ureas, triazines, carbamates or diphenyl ethers.

EP-A-0 558 448 describes 1,5-diphenylpyrazole-3-carboxylic acid derivatives for protecting against the phytotoxic effect of sulfonylureas.

It has now been found that safener compounds selected from the classes of the quinoline, pyrazole or triazole derivatives are suitable for protecting crop plants against the phytotoxic effect of 4-aryl- and 4-heteroaryl-5-oxopyrazoline derivatives of the formula I.

In accordance with the invention, therefore, a selective-herbicidal composition is proposed which comprises, in addition to customary inert formulation auxiliaries such as carriers, solvents and wetting agents, an active substance comprising a mixture of

- a) a herbicidally effective quantity of a herbicide compound of the formula I as defined hereinbefore and
- b) a herbicide-antagonistically effective quantity of either a quinoline derivative of the formula

$$\begin{array}{c}
X_2 \\
O-CH_2-C-O-R_{15}
\end{array}$$
(X),

in which

R₁₅ is hydrogen, C₁-C₈alkyl or C₁-C₈alkyl which is substituted by C₁-C₆alkoxy or

C₃-C₆alkenyloxy; and

X₂ is hydrogen or chlorine; or

of a 1-phenylazole-3-carboxylic acid derivative of the formula

$$R_{16}$$

$$R_{16}$$

$$R_{17}$$

$$R_{18}$$
(XI),

in which

E is nitrogen or methine;

R₁₆ is -CCl₃ or unsubstituted or halo-substituted phenyl;

 R_{17} and R_{18} independently of one another are hydrogen or halogen; and R_{19} is C_1 - C_4 alkyl.

Preferred compounds of the formula I in the composition according to the invention are those in which R_1 is the group $(R_4)_n$; and m is 0. Among these, particular

preference is given to those compounds in which R_4 is C_1 - or C_2 alkyl; and n is 1, 2 or 3.

In particular, among these compounds, very particular preference is given to those in which R_4 is C_1 - or C_2 alkyl in positions 2, 4 and 6 relative to the phenyl linkage site; and n is 3.

Likewise preferred are compositions according to the invention in which, in the

compounds of the formula I, R_1 is the group $(R_4)_n$; m is 0; R_4 is C_1 - or

 C_2 alkyl; n is 1, 2 or 3; R_2 and R_3 are C_1 - C_6 alkyl; or R_2 and R_3 together form a substituted or unsubstituted alkylene chain -(CH_2)₅-, -(CH_2)₄- or -(CH_2)₃-.

Also preferred are those compositions in which G is the group -CO-A (a); and A is C_1 - C_8 alkyl substituted by nitro, cyano, $Si(C_1$ - C_4 alkyl)₃, amino, C_1 - C_4 alkylamino, $di(C_1$ - C_4 alkyl)amino, unsubstituted or substituted benzyloxy or a group $-\frac{CH-NR_{08}R_{09}}{COOR_{010}}$,

or A is C_3 - C_8 cycloalkyl which if desired can contain as heteroatom oxygen, sulfur or nitrogen and is substituted by C_1 - C_4 alkyl, C_1 - C_4 alkoxy, C_1 - C_4 alkylthio, C_1 - C_4 alkylsulfinyl, C_1 - C_4 alkylsulfonyl or substituted or unsubstituted phenyl, or A is C_3 - C_8 cycloalkyloxy which can if desired contain as heteroatom oxygen, sulfur or nitrogen and is unsubstituted or substituted by C_1 - C_4 alkyl, or A is a group - $(CR_8R_9)_p$ - $(Y)_o$ -CO- $(Z)_r$ - R_{10} or - $(CR_8R_9)_p$ - R_{11} .

Furthermore, preference is given to those compositions in which R_1 is the group $\binom{(R_4)_n}{(R_5)_m}$; R_4 is C_1 - or C_2 alkyl; n is 1, 2 or 3; m is 0; R_2 and R_3 are C_1 - C_6 alkyl; or

 R_2 and R_3 together form a substituted or unsubstituted alkylene chain -(CH₂)₅-, -(CH₂)₄- or -(CH₂)₃-; G is the group -CO-A (a); and A is C₁-C₈alkyl substituted by nitro, cyano, Si(C₁-C₄alkyl)₃, amino, C₁-C₄alkylamino, di(C₁-C₄alkyl)amino, unsubstituted or substituted benzyloxy or a group $\frac{-CH-NR_{08}R_{09}}{COOR_{010}}$, or A is C₃-C₈cycloalkyl which can if $\frac{COOR_{010}}{COOR_{010}}$

desired contain as heteroatom oxygen, sulfur or nitrogen and is substituted by C_1 - C_4 alkyl, C_1 - C_4 alkoxy, C_1 - C_4 alkylthio, C_1 - C_4 alkylsulfinyl, C_1 - C_4 alkylsulfonyl or substituted or unsubstituted phenyl, or A is C_3 - C_8 cycloalkyloxy which can if desired contain as heteroatom oxygen, sulfur or nitrogen and is unsubstituted or substituted by C_1 - C_4 alkyl, or A is a group - $(CR_8R_9)_p$ - $(Y)_o$ -CO- $(Z)_r$ - R_{10} or - $(CR_8R_9)_p$ - R_{11} .

Furthermore, preference is given to those compositions which comprise a safener of the formula

Likewise preferred are compositions which comprise a safener of the formula

Further preferred compositions are those which comprise a safener of the formula

Further preferred compounds of the formulae X and XI are also listed in Tables 8 and 9.

The quinoline derivatives from the scope of the formula X and their preparation are known or they can be prepared by analogy with known processes, as described for example in EP-A-0 094 349.

The 1-phenylazole-3-carboxylic acid derivatives from the scope of the formula XI are known and are described for example in EP-A-0 268 554 and EP-A-0 174 562.

In the field of pest control the compounds of the formula I according to the invention, used even at low concentrations, are active substances having preventive and/or curative value, a very favourable biocidal spectrum and favourable tolerance by warm-blooded animals, fish and plants. The active substances according to the invention are effective against all or individual development stages of normally sensitive, but also of resistant, animal pests,

such as insects or members of the order Acarina. The insecticidal or acaricidal action of the active substances according to the invention can become apparent directly, i.e. in destruction of the pests which occurs immediately or only after a certain time, for example during moulting, or indirectly, for example in reduced oviposition and/or hatching rates, the favourable action corresponding to a rate of destruction (mortality) of at least 50 to 60 %.

The animal pests mentioned include, for example:

from the order Lepidoptera, for example

Acleris spp., Adoxophyes spp., Aegeria spp., Agrotis spp., Alabama argillaceae, Amylois spp., Anticarsia gemmatalis, Archips spp., Argyrotaenia spp., Autographa spp., Busseola fusca, Cadra cautella, Carposina nipponensis, Chilo spp., Choristoneura spp., Clysia ambiguella, Cnaphalocrocis spp., Cnephasia spp., Cochylis spp., Coleophora spp., Crocidolomia binotalis, Cryptophlebia leucotreta, Cydia spp., Diatraea spp., Diparopsis castanea, Earias spp., Ephestia spp., Eucosma spp., Eupoecilia ambiguella, Euproctis spp., Euxoa spp., Grapholita spp., Hedya nubiferana, Heliothis spp., Hellula undalis, Hyphantria cunea, Keiferia lycopersicella, Leucoptera scitella, Lithocollethis spp., Lobesia botrana, Lymantria spp., Lyonetia spp., Malacosoma spp., Mamestra brassicae, Manduca sexta, Operophtera spp., Ostrinia nubilalis, Pammene spp., Pandemis spp., Panolis flammea, Pectinophora gossypiella, Phthorimaea operculella, Pieris rapae, Pieris spp., Plutella xylostella, Prays spp., Scirpophaga spp., Sesamia spp., Sparganothis spp., Spodoptera spp., Synanthedon spp., Thaumetopoea spp., Tortrix spp., Trichoplusia ni and Yponomeuta spp.;

from the order Coleoptera, for example

Agriotes spp., Anthonomus spp., Atomaria linearis, Chaetocnema tibialis, Cosmopolites spp., Curculio spp., Dermestes spp., Diabrotica spp., Epilachna spp., Eremnus spp., Leptinotarsa decemlineata, Lissorhoptrus spp., Melolontha spp., Orycaephilus spp., Otiorhynchus spp., Phlyctinus spp., Popillia spp., Psylliodes spp., Rhizopertha spp., Scarabeidae, Sitophilus spp., Sitotroga spp., Tenebrio spp., Tribolium spp. and Trogoderma spp.;

from the order Orthoptera, for example

Blatta spp., Blattella spp., Gryllotalpa spp., Leucophaea maderae, Locusta spp.,

Periplaneta spp. and Schistocerca spp.;

from the order Isoptera, for example,

Reticulitermes spp.;

from the order Psocoptera, for example

Liposcelis spp.;

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from the order Anoplura, for example

Haematopinus spp., Linognathus spp., Pediculus spp., Pemphigus spp. and Phylloxera spp.;

from the order Mallophaga, for example

Damalinea spp. and Trichodectes spp.;

from the order Thysanoptera, for example

Frankliniella spp., Hercinothrips spp., Taeniothrips spp., Thrips palmi, Thrips tabaci and Scinothrips aurantii;

from the order Heteroptera, for example

Cimex spp., Distantiella theobroma, Dysdercus spp., Euchistus spp., Eurygaster spp.

Leptocorisa spp., Nezara spp., Piesma spp., Rhodnius spp., Sahlbergella singularis,

Scotinophara spp. and Triatoma spp.;

from the order Homoptera, for example

Aleurothrixus floccosus, Aleyrodes brassicae, Aonidiella spp., Aphididae, Aphis spp.,

Aspidiotus spp., Bemisia tabaci, Ceroplaster spp., Chrysomphalus aonidium,

Chrysomphalus dictyospermi, Coccus hesperidum, Empoasca spp., Eriosoma larigerum,

Erythroneura spp., Gascardia spp., Laodelphax spp., Lecanium corni, Lepidosaphes spp.,

Macrosiphus spp., Myzus spp., Nephotettix spp., Nilaparvata spp., Paratoria spp.,

Pemphigus spp., Planococcus spp., Pseudaulacaspis spp., Pseudococcus spp., Psylla spp.,

Pulvinaria aethiopica, Quadraspidiotus spp., Rhopalosiphum spp., Saissetia spp.,

Scaphoideus spp., Schizaphis spp., Sitobion spp., Trialeurodes vaporariorum, Trioza erytreae and Unaspis citri;

from the order Hymenoptera, for example

Acromyrmex, Atta spp., Cephus spp., Diprion spp., Diprionidae, Gilpinia polytoma, Hoplocampa spp., Lasius spp., Monomorium pharaonis, Neodiprion spp., Solenopsis spp. and Vespa spp.;

from the order Diptera, for example

Aedes spp., Antherigona soccata, Bibio hortulanus, Calliphora erythrocephala, Ceratitis spp., Chrysomyia spp., Culex spp., Cuterebra spp., Dacus spp., Drosophila melanogaster, Fannia spp., Gastrophilus spp., Glossina spp., Hypoderma spp., Hyppobosca spp., Liriomyza spp., Lucilia spp., Melanagromyza spp., Musca spp., Oestrus spp., Orseolia spp., Oscinella frit, Pegomyia hyoscyami, Phorbia spp., Rhagoletis pomonella, Sciara spp., Stomoxys spp., Tabanus spp., Tannia spp. and Tipula spp.;

from the order Siphonaptera, for example

Ceratophyllus spp. and Xenopsylla cheopis;

from the order Thysanura, for example

Lepisma saccharina and

from the order Acarina, for example

Acarus siro, Aceria sheldoni, Aculus schlechtendali, Amblyomma spp., Argas spp., Boophilus spp., Brevipalpus spp., Bryobia praetiosa, Calipitrimerus spp., Chorioptes spp., Dermanyssus gallinae, Eotetranychus carpini, Eriophyes spp., Hyalomma spp., Ixodes spp., Olygonychus pratensis, Ornithodoros spp., Panonychus spp., Phyllocoptruta oleivora, Polyphagotarsonemus latus, Psoroptes spp., Rhipicephalus spp., Rhizoglyphus spp., Sarcoptes spp., Tarsonemus spp. and Tetranychus spp..

Using the active substances according to the invention it is possible to control pests of the type mentioned which occur on plants, especially on useful and ornamental plants in agriculture, gardening and forestry, or on parts of such plants, such as fruits, flowers, foliage, stems, tubers or roots, control in this context meaning restrict or destroy, while in some cases also providing protection against these pests to parts of plants which grow subsequently.

Suitable target crops are, in particular, cereals such as wheat, barley, rye, oats, rice, maize or sorghum; beet, such as sugar and fodder beet; fruit, for example pome fruit, stone fruit and soft fruit, such as apples, pears, plums, peaches, almonds, cherries or berries, for example strawberries, raspberries or blackberries; pulses, such as beans, lentils, peas or soya; oil crops, such as oilseed rape, mustard, poppy, olives, sunflowers, coconut, castor, cacao or groundnuts; cucurbits, such as pumpkins, cucumbers or melons; fibre plants such as cotton, flax, hemp or jute; citrus fruits, such as oranges, lemons, grapefruits or tangerines; vegetables, such as spinach, lettuce, asparagus, cabbages, carrots, onions, tomatoes, potatoes or bell pepper; the laurel family, such as avocado, cinnamon or camphor; and also tobacco, nuts, coffee, eggplants, sugar cane, tea, pepper, vines, hops, banana family, latex plants and ornamentals.

Further fields of use of the active substances according to the invention are the protection of stocks, warehouses and material and in the hygiene sector, especially the protection of domestic and productive animals against pests of the type mentioned.

The invention therefore also relates to pesticides which are forms, to be chosen in accordance with the intended aims and prevailing conditions, such as emulsifiable concentrates, suspension concentrates, directly sprayable or dilutable solutions, brushable pastes, dilute emulsions, wettable powders, soluble powders, dispersible powders, dusts, granules or encapsulations in polymeric substances, and which comprise - at least - one of the active substances according to the invention.

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The active substance is employed in these compositions in pure form, a solid active substance, for example, in a specific particle size, or, preferably, together with - at least - one of the auxiliaries which are customary in the art of formulation, such as extenders, for example solvents or solid carriers, or surface-active compounds (surfactants).

Examples of suitable solvents are unmodified or partially hydrogenated aromatic hydrocarbons, preferably the C₈ to C₁₂ fractions of alkylbenzenes, such as xylene mixtures, alkylated naphthalenes or tetrahydronaphthalene, aliphatic or cycloaliphatic hydrocarbons, such as paraffins or cyclohexane, alcohols, such as ethanol, propanol or butanol, glycols and ethers and esters thereof, such as propylene glycol, dipropylene glycol ether, ethylene glycol or ethylene glycol monomethyl or monoethyl ether, ketones, such as cyclohexanone, isophorone or diacetone alcohol, strongly polar solvents, such as N-methylpyrrolid-2-one, dimethyl sulfoxide or N,N-dimethylformamide, water, unmodified or epoxidized vegetable oils, such as unmodified or epoxidized rapeseed, castor, coconut or soya oil, and silicone oils.

As solid carriers, for example for dusts and dispersible powders, use is generally made of ground natural minerals, such as calcite, talc, kaolin, montmorillonite or attapulgite. To improve the physical properties it is also possible to add highly disperse silicas or highly disperse absorbent polymers. Suitable granulated, adsorptive granule carriers are porous types, such as pumice, broken brick, sepiolite or bentonite, while suitable non-sorbent carrier materials are calcite or sand. Moreover, a large number of granulated materials of inorganic or organic type, especially dolomite or comminuted plant residues, can be used.

Suitable surface-active compounds, depending on the nature of the active substance to be formulated, are nonionic, cationic and/or anionic surfactants, or surfactant mixtures having good emulsifying, dispersing and wetting properties. The surfactants listed below should be regarded only as examples; the relevant literature describes many other surfactants which are common in the art of formulation and are suitable in accordance with the invention.

Suitable nonionic surfactants are primarily polyglycol ether derivatives of aliphatic or cycloaliphatic alcohols, saturated or unsaturated fatty acids and alkylphenols, which can have 3 to 30 glycol ether groups and 8 to 20 carbon atoms in the (aliphatic) hydrocarbon radical and 6 to 18 carbon atoms in the alkyl radical of the alkylphenols. Also suitable are water-soluble adducts, containing 20 to 250 ethylene glycol ether groups and 10 to 100

propylene glycol ether groups, of polyethylene oxide with polypropylene glycol, ethylenediaminopolypropylene glycol and alkylpolypropylene glycol having 1 to 10 carbon atoms in the alkyl chain. The compounds mentioned usually contain, per propylene glycol unit, 1 to 5 ethylene glycol units. Examples which may be mentioned are nonylphenolpolyethoxyethanols, castor polyglycol ethers, polypropylene-polyethylene oxide adducts, tributylphenoxypolyethoxyethanol, polyethylene glycol and octylphenoxypolyethoxyethanol. Also suitable are fatty acid esters of polyoxyethylenesorbitan, such as polyoxyethylenesorbitan trioleate.

The cationic surfactants are primarily quaternary ammonium salts which have as substituents at least one alkyl radical having 8 to 22 carbon atoms and, additionally, lower, unmodified or halogenated, alkyl, benzyl or lower hydroxyalkyl radicals. These salts are preferably halides, methyl sulfates or ethyl sulfates. Examples are stearyltrimethylammonium chloride and benzyldi(2-chloroethyl)ethylammonium bromide.

Suitable anionic surfactants can be both water-soluble soaps and water-soluble synthetic surface-active compounds. Suitable soaps are the alkali metal, alkaline earth metal and substituted or unsubstituted ammonium salts of higher fatty acids $(C_{10}-C_{22})$, such as the sodium or potassium salts of oleic or stearic acid, or of natural fatty acid mixtures which can be obtained, for example, from coconut oil or tall oil; the fatty acid methyl-taurine salts should also be mentioned. More often, however, synthetic surfactants are used, especially fatty sulfonates, fatty sulfates, sulfonated benzimidazole derivatives or alkylarylsulfonates. The fatty sulfonates and fatty sulfates are generally in the form of alkali metal, alkaline earth metal or substituted or unsubstituted ammonium salts and generally have an alkyl radical of 8 to 22 carbon atoms, alkyl also including the alkyl moiety of acyl radicals; examples which may be mentioned are the sodium or calcium salt of ligninsulfonic acid, of dodecylsulfuric ester or of a fatty alcohol sulfate mixture prepared from natural fatty acids. Also included are the salts of sulfuric esters and sulfonic acids of fatty alcohol-ethylene oxide adducts. The sulfonated benzimidazole derivatives preferably contain 2 sulfonic acid groups and a fatty acid radical having about 8 to 22 carbon atoms. Alkylarylsulfonates are, for example, the sodium, calcium or triethanolammonium salts of dodecylbenzenesulfonic acid, of dibutylnaphthalenesulfonic acid or of a naphthalenesulfonic acid-formaldehyde condensation product. Also suitable are appropriate phosphates, such as salts of the phosphoric ester of a p-nonylphenol-(4-14)-ethylene oxide adduct or phospholipids.

The compositions generally contain from 0.1 to 99 %, in particular from 0.1 to 95 %, of

active substance and from 1 to 99.9 %, in particular from 5 to 99.9 % of - at least - one solid or liquid auxiliary, it being possible in general for from 0 to 25 %, in particular from 0.1 to 20 %, of the compositions to be surfactants (% denotes in each case percent by weight). Whereas the commercial product tends preferably to comprise concentrated compositions, the end user generally uses dilute compositions with substantially lower concentrations of active substance. Preferred compositions are composed, in particular, as follows (% = percent by weight):

Emulsifiable concentrates:

Active substance: from 1 to 90%, preferably from 5 to 20%

Surfactant: from 1 to 30%, preferably from 10 to 20%

Solvent: from 5 to 98%, preferably from 70 to 85%

Dusts:

Active substance: from 0.1 to 10%, preferably from 0.1 to 1%

Solid carrier: from 99.9 to 90%, preferably from 99.9 to 99%

Suspension concentrates:

Active substance: from 5 to 75%, preferably from 10 to 50%

Water: from 94 to 24%, preferably from 88 to 30%

Surfactant: from 1 to 40%, preferably from 2 to 30%

Wettable powders:

Active subtance: from 0.5 to 90%, preferably from 1 to 80%

Surfactant: from 0.5 to 20%, preferably from 1 to 15%

Solid carrier: from 5 to 99%, preferably from 15 to 98%

Granules:

Active substance: from 0.5 to 30%, preferably from 3 to 15%

Solid carrier: from 99.5 to 70%, preferably from 97 to 85%

The action of the compositions according to the invention can be substantially broadened and adapted to prevailing circumstances by the addition of other insecticidal or acaricidal active substances. In this context, suitable additions of insecticidal or acaricidal active substance are, for example, representatives of the following classes of active substance: organophosphorus compounds, nitrophenols and derivatives, formamidines, acylureas, carbamates, pyrethroids, nitroenamines and derivatives, pyrroles, thioureas and

derivatives, chlorinated hydrocarbons and Bacillus thuringiensis preparations. The compositions according to the invention can also comprise further solid or liquid auxiliaries, such as stabilizers, for example unmodified or epoxidized vegetable oils (e.g. epoxidized coconut oil, rapeseed oil or soya oil), defoamers, for example silicone oil, preservatives, viscosity regulators, binders and/or adhesives, and also fertilizers or other active substances for obtaining specific effects, examples being bactericides, nematicides, molluscicides or selective herbicides.

The compositions according to the invention are prepared in a known manner: for example in the absence of auxiliaries, by grinding, sieving and/or compression of a solid active substance or active substance mixture, for example to a defined particle size, and, when at least one auxiliary is present, for example by intimate mixing and/or grinding of the active substance or active substance mixture with the auxiliary or auxiliaries. These processes for producing the compositions according to the invention, and the use of the compounds I for the production of these compositions, are likewise a subject of the invention.

The invention relates furthermore to the methods of application for the compositions, i.e. the methods of controlling pests of the type mentioned, these methods being chosen in accordance with the intended aims and prevailing circumstances and comprising, for instance, spraying, fogging, dusting, brushing, dressing, broadcasting or watering, and to the use of the compositions for controlling pests of the type mentioned. In this context, typical application concentrations are between 0.1 and 1000 ppm, preferably between 0.1 and 500 ppm, of active substance. The application rates per hectare are generally from 1 to 200 g of active substance per hectare, in particular from 10 to 1000 g/ha, preferably from 20 to 600 g/ha.

A preferred method of application in the area of plant protection is application to the foliage of the plants (foliar application), the frequency and rate of application depending on the extent of infestation by the respective pest. However, the active substance can also pass into the plants via the root system (systemic action), by saturating the site of the plants with a liquid composition or incorporating the active substance in solid form into the site of the plants, for example into the soil, in the form of granules, for example (soil application). In paddy rice cultures, such granules can be metered out into the flooded rice field.

The compositions according to the invention are also suitable for protecting vegetative propagation stock, for example seeds, such as fruits, tubers or grains, or plant seedlings,

against animal pests. In this case the propagation stock can be treated with the composition before being planted out; seed, for example, can be dressed before sowing. The active substances according to the invention can also be applied to seeds (coating) by either soaking the seeds in a liquid composition or coating them with a solid composition. The composition can also be applied when the propagation stock is planted out, at the planting site, for example in the seed drill in the case of sowing. These treatment methods for vegetative propagation stock, and the vegetative propagation stock thus treated, are further subjects of the invention.

The compounds of the formula I according to the invention are also herbicidally active. Suitable application methods for the herbicidal use, according to the invention, of the compounds of the formula I or of compositions comprising them are all those methods which are customary in agriculture, such as preemergence application, postemergence application and seed dressing, and also a variety of methods and techniques such as, for example, the controlled release of active substance. For this purpose, the active substance in solution is applied to mineral granular carriers or polymerized granules (urea/formaldehyde) which are then dried. If desired, it is additionally possible to apply a coating (coated granules) which permits the release of the active substance in regulated amounts over a defined period.

The compounds of the formula I can be employed in unaltered form, i.e. as they are obtained in the synthesis, but are preferably processed in a conventional manner, using the auxiliaries which are customary in the art of formulation, for example to give emulsifiable concentrates, directly sprayable or dilutable solutions, dilute emulsions, wettable powders, soluble powders, dusts, granules or microcapsules. The methods of application, such as spraying, misting, dusting, wetting, scattering or pouring, are - like the nature of the compositions - chosen in accordance with the intended aims and the prevailing conditions.

The formulations, i.e. the compositions, preparations or combinations comprising the active substance of the formula I or at least one active substance of the formula I and, if desired, one or more solid or liquid additives, are produced in a known manner, for example by intimate mixing and/or grinding of the active substances with the additives, such as solvents or solid carriers, for example. It is also possible, in addition, for surface-active compounds (surfactants) to be used in the production of the formulations. Suitable solvents, solid carriers and surface-active compounds are, for example, of the type described hereinbefore.

The herbicidal formulations generally contain from 0.1 to 99 %, in particular from 0.1 to

95 %, of active substance of the formula I, from 1 to 99 % of a solid or liquid formulation auxiliary and from 0 to 25 %, in particular from 0.1 to 25 %, of a surfactant.

While the commercial product tends preferably to comprise concentrated compositions, the end user generally uses dilute compositions.

The compositions can also comprise further additives, such as stabilizers, for example unmodified or epoxidized vegetable oils (epoxidized coconut oil, rapeseed oil or soya oil), defoamers, for example silicone oil, preservatives, viscosity regulators, binders, adhesives, and also fertilizers or other active substances for obtaining specific effects.

In particular, preferred formulations are composed as follows (% = percent by weight):

Emulsifiable concentrates:

Active substance:

from 1 to 90%, preferably from 5 to 50%

Surfactant:

from 5 to 30%, preferably from 10 to 20%

Solvent:

from 15 to 94%, preferably from 70 to 85%

Dusts:

Active substance:

from 0.1 to 50%, preferably from 0.1 to 1%

Solid carrier:

from 99.9 to 90%, preferably from 99.9 to 99%

Suspension concentrates:

Active substance:

from 5 to 75%, preferably from 10 to 50%

Water:

from 94 to 24%, preferably from 88 to 30%

Surfactant: from 1 to 40%

from 1 to 40%, preferably from 2 to 30%

Wettable powders:

Active subtance:

from 0.5 to 90%, preferably from 1 to 80%

Surfactant:

from 0.5 to 20%, preferably from 1 to 15%

Solid carrier:

from 5 to 95%, preferably from 15 to 90%

Granules:

Active substance:

from 0.1 to 30%, preferably from 0.1 to 15%

Solid carrier:

from 99.5 to 70%, preferably from 97 to 85%

The active substances of the formula I are in general employed successfully at application

rates of from 0.001 to 2 kg/ha, in particular from 0.005 to 1 kg/ha. The rate required in order to achieve the desired effect can be determined by means of trials. It is dependent on the nature of the effect, on the stage of development of the crop plant and of the weed, and on the application (site, time, method) and can, depending on these parameters, vary within wide ranges.

The compounds of the formula I are distinguished by herbicidal properties which render them outstanding for use in crops of useful plants, especially in cereals, cotton, soya, oilseed rape, maize, rice and plantation crops. They can additionally be employed as burn-down agents, for example for potatoes, or as defoliants.

References to cultures or crops are intended to include those which have been given tolerance to herbicides or classes of herbicide by means of conventional breeding methods or methods of genetic manipulation.

The invention also relates to a process for the selective control of broad-leaved weeds and grasses in crops of useful plants, which comprises treating the useful plants, their seeds or seedlings or their cultivation area, simultaneously or independently of one another, with a herbicidally effective quantity of the herbicide of the formula I and with a herbicide-antagonistically effective quantity of a safener of the formula X or XI.

Crop plants which can be protected by the safeners of the formulae X and XI against the damaging effect of the abovementioned herbicides are, in particular, maize and cereals.

The weeds to be controlled can be either monocotyledon or dicotyledon weeds.

The areas regarded as cultivation areas are the soil areas on which the crop plants are already growing, or soil areas sown with the seed of these crop plants, and also the areas intended for cultivation with these crop plants.

Depending on the intended use, a safener of the formula X or XI can be used for pretreating the seed of the crop plant (dressing of the seed or of the seedlings), or placed in the soil before or after sowing. It can also, however, be applied, on its own or together with the herbicide, following the emergence of the plants. The treatment of the plants or of the seed with the safener can therefore, in principle, take place independently of the point in time of the application of the herbicide. The plant treatment, however, can also be carried out by simultaneous application of herbicide and safener (for example as a tank

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mix).

The application rate of safener to herbicide to be applied depends largely on the method of application. In a field treatment carried out either using a tank mix comprising a combination of safener and herbicide or by separate application of safener and herbicide, the ratio of herbicide to safener is in general from 100:1 to 1:10, preferably from 20:1 to 1:1.

In general, in field treatment, from 0.001 to 5.0 kg of safener/ha, preferably from 0.001 to 0.5 kg of safener/ha, are applied.

The application rates of herbicide are generally between 0.001 and 2 kg/ha, but preferably between 0.005 and 1 kg/ha.

The compositions according to the invention are suitable for all application methods which are customary in agriculture, for example preemergence application, postemergence application and seed dressing.

In the case of seed dressing, in general, from 0.001 to 10 g of safener/kg of seed, preferably from 0.05 to 2 g of safener/kg of seed, are applied. If the safener is applied in liquid form shortly before sowing, accompanied by seed swelling, then it is expedient to use safener solutions which comprise the active substance in a concentration of from 1 to 10 000 ppm, preferably from 100 to 1000 ppm.

For application, the safeners of the formula X or XI, or combinations of these safeners with the herbicides of the formula I, are advantageously processed together with the auxiliaries which are customary in the art of formulation to give particular formulations, for example emulsion concentrates, brushable pastes, directly sprayable or dilutable solutions, dilute emulsions, wettable powders, soluble powders, dusts, granules or microcapsules. The formulations are prepared in a known manner, for example by intimate mixing and/or grinding of the active substances with liquid or solid formulation auxiliaries such as, for example, solvents or solid carriers. Furthermore, it is possible in addition to use surface-active compounds (surfactants) when producing the formulations. Suitable solvents, solid carriers and surface-active compounds are, for example, of the type described hereinbefore.

The agrochemical formulations generally contain from 0.1 to 99 percent by weight, in

particular from 0.1 to 95 % by weight, of safener or safener/herbicide active-substance mixture, from 1 to 99 % by weight, in particular from 5 to 99.8 % by weight, of a solid or liquid formulation auxiliary, and from 0 to 25 % by weight, in particular from 0.1 to 25 % by weight, of surfactant.

While the preferred commercial product usually comprises concentrated compositions, the end user generally uses dilute compositions.

The compositions can also comprise further additives, such as stabilizers, defoamers, viscosity regulators, binders, adhesives and fertilizers or other active substances.

For the use of safeners of the formulae X and XI or compositions comprising them for protecting crop plants against the damaging effects of herbicides of the formula I, a variety of methods and techniques are suitable, examples being the following:

i) Seed dressing

- a) The seeds are dressed by shaking them, in a vessel, together with an active substance of the formula X or XI formulated as a wettable powder until the active ingredient is distributed uniformly on the seed surface (dry seed dressing). In this case from about 1 to about 500 g of active substance of the formula X or XI (from 4 g to 2 kg of wettable powder) are used per 100 kg of seed.
- b) The seeds are dressed with an emulsion concentrate of the active substance of the formula X or XI in accordance with method a) (wet seed dressing).
- c) The seeds are dressed by immersing them into a liquor containing 100-1000 ppm of active substance of the formula X or XI for from 1 to 72 hours and then, if desired, by drying them (seed soaking).

The dressing of the seed, and the treatment of the seedling which has begun germinating, are of course the preferred methods of application, since the treatment with the active substance is directed entirely towards the target crop. As a rule, from 1 to 1000 g of antidote, preferably from 5 to 250 g of antidote, are used per 100 kg of seed, it being possible to deviate from these limit concentrations up or down repeat dressing, depending on the method, which also makes possible the addition of other active substances or micronutrients.

ii) Application as a tank mix

A liquid preparation of a mixture of antidote and herbicide (mutual quantitative ratio of between 10:1 and 1:100) is used, the application rate of herbicide being from 0.005 to 5.0 kg per hectare. Such tank mixes are applied before or after sowing.

iii) Application in the seed drill

The safener is incorporated into the open seed drill, containing seed, as an emulsion concentrate, wettable powder or in the form of granules. After the seed drill has been covered over, the herbicide is applied preemergence in a customary manner.

iv) Controlled release of active substance

The active substance of the formula X or XI is applied in solution to mineral granule carriers or polymerized granules (urea/formaldehyde), which are then dried. If desired, a coating can be applied (coated granules), which enables the release of the active substance in regulated amounts over a defined period.

The examples which follow illustrate the invention in more detail without limiting it. Temperatures are given in degrees Celsius. The temperatures given in the Tables, in the column "Phys. data", are the melting points of the corresponding compounds unless otherwise indicated.

Preparation Examples:

Example H1: Preparation of 3-oxo-2-(2,4,6-trimethylphenyl)-5,6,7,8-tetrahydro-3-H-pyrazolo[1,2-a]pyridazin-1-yl 2-cyclohexyl-2-methylpropionate

$$\begin{array}{c|c}
C & CH_3 \\
C & CH_3
\end{array}$$

$$\begin{array}{c|c}
C & CH_3
\end{array}$$

1.09 g of 3-hydroxy-4-mesityl-5-oxo-1,2-tetramethylenepyrazoline and 1.4 ml of triethylamine are dissolved in 25 ml of tetrahydrofuran, and 0.75 g of 2-cyclohexyl-2-methylpropionic acid is added at 20°. The reaction mixture is stirred at 40° for 3 hours and then concentrated on a rotary evaporator. The residue is taken up in toluene and water. The toluene phase is dried over sodium sulfate and concentrated. The residue is stirred in

hexane and then filtered off with suction to give the desired compound as a white powder having a melting point of 172-173°.

Example H2: Preparation of 3-oxo-2-(2,4,6-trimethylphenyl)-5,6,7,8-tetrahydro-3-H-pyrazolo[1,2-a]pyridazin-1-yl (2-cyanoethyl)methylcarbamate

$$CH_3$$
 CH_2CH_2CN
 CH_3
 C

1.22 g of 3-hydroxy-4-mesityl-5-oxo-1,2-tetramethylenepyrazoline and 1.14 g of triethylamine are dissolved in 25 ml of tetrahydrofuran, and 0.7 g of (2-cyanoethyl)-methylcarbamoyl chloride is added with stirring. The reaction mixture is stirred at 40° for 3 hours and then concentrated on a rotary evaporator. Dichloromethane and ice-water are added to the residue. The organic phase is dried with sodium sulfate and concentrated, and the residue obtained is stirred with hexane, filtered off with suction and dried to give the desired compound as a white powder with a melting point of 151-153°.

Example H3: Preparation of 3-oxo-2-(2,4,6-trimethylphenyl)-5,6,7,8-tetrahydro-3H-pyrazolo[1,2-a]pyridazin-1-yl cyclohexylmethylsulfamate

2.7 g of 3-hydroxy-4-mesityl-5-oxo-1,2-tetramethylenepyrazoline and 3.0 g of triethylamine are dissolved in 30 ml of tetrahydrofuran, a solution of 2.1 g of cyclohexylmethylsulfamoyl chloride is added at room temperature, and the reaction mixture is then stirred at 40° for 8 hours. It is concentrated and the residue is taken up with tert-butyl methyl ether and ice-water. The organic phase is dried with sodium sulfate and concentrated. The crude product is dissolved in ethyl acetate, the solution is filtered over silica gel, the filtrate is concentrated and the residue is stirred with hexane. Suction filtration and drying give the desired compound as a white powder with a melting point of

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171-173°.

The compounds of Tables 1-9 can be prepared by analogy with the above examples and with the preparation methods described.

Tables 1-7 below list, as compounds of the formulae Ia to Ig, compounds of the formula I which are preferred as insecticides and acaricides and also as herbicides.

Tables 8 and 9 list compounds which are preferred as safeners of the formulae X and XI.

$$\begin{array}{c|c}
 & O \\
 & \parallel \\
 & C \\$$

Comp.	R ₂ R ₃	A	Phys.data
1.1	-(CH ₂) ₄ -	-CH ₂ FF	125-127°
1.2	-(CH ₂) ₄ -	-CH ₂ -(CH ₂) ₇	65-67°
1.3	-(CH ₂) ₄ -	-CH ₂ CH ₂ -F	103-106°
1.4	-(CH ₂) ₄ -	-CH ₂ CH ₂	111-113°
1.5	-(CH ₂) ₄ -	-CH ₂ —	106-108°
1.6	-(CH ₂) ₄ -	-CH ₂ CH ₂ —	102-104°
1.7	-(CH ₂) ₄ -	-CH ₂	80-82°
1.8	-(CH ₂) ₄ -	-N(CH ₃)CH ₂ CH ₂ CN	151-153°
1.9	-(CH ₂) ₄ -	-CH ₂	101-103°
1.10	-(CH ₂) ₄ -	-N(CH ₂ CH ₂ CN)-(CH ₂) ₃ OC ₂ H ₅	
1.11	-(CH ₂) ₄ -	-CH-CH-CH-	90-92°

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Comp No.	. R ₂ R ₃	A	Phys.data
1.12	-(CH ₂) ₄ -	-CH ₂ O	150-152°
1.13	-(CH ₂) ₄ -	-CH(C ₆ H ₅)OCOCH ₃	145-146°
1.14	-(CH ₂) ₄ -	-C(CH ₃) ₂ OCOCH ₃	104°
1.15	-(CH ₂) ₄ -		179-180°
1.16	-(CH ₂) ₄ -		172-173°
1.17	-CH ₂ -CH-CH-CH ₂ - CH ₃ CH ₃	-CH ₂	137-139°
1.18	-(CH ₂) ₄ -	-C(CH ₃) ₂	172-173°
1.19	-(CH ₂) ₄ -	adamant-1-yl	204-205°
1.20	-CH ₂ -CH-CH-CH ₂ - CH ₃ CH ₃	-C(CH ₃) ₂	134-136°
1.21	-(CH ₂) ₄ -		120-121°
1.22	CH ₃ CH ₃	-C(CH ₃) ₂ OCOCH ₃	148-149°

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Comp No.	. R ₂ R ₃	A	Phys.data
1.23	CH ₃ CH ₃		135-136°
1.3.1 · 电电池调整:	gradient generalist in the second second	and the state of t	
1.24	-(CH ₂) ₄ -		amorphous
1.25	-(CH ₂) ₄ -	-CH ₂ CH ₂ CH ₂ COOC ₂ H ₅	resin
1.26	-(CH ₂) ₄ -	-C(CH ₃) ₂ OCO-t-C ₄ H ₉	149-1 50°
1.27	-(CH ₂) ₄ -	-C(CH ₃) ₂ OCO-C ₆ H ₅	143-145°
1.28	-(CH ₂) ₄ -	-N(CH ₃)—	resin
1.29	CH ₃ CH ₃	-C(CH ₃) ₂	160-161°
1.30	-(CH ₂) ₄ -	-CH ₂ Si(CH ₃) ₃	
1.31	-(CH ₂) ₄ -	X	•
1.32	-(CH ₂) ₄ -	-C(CH ₃) ₂ COOCH ₃	
1.33	-(CH ₂) ₄ -	CH ₃	135-137°
1.34	-(CH ₂) ₄ -	Z ^{C₆H₅}	120°
1.35	-(CH ₂) ₄ -		oil
1.36	-(CH ₂) ₃ -	$-C(CH_3)_2OC(=O)CH_3$	resin
1.37	-CH ₂ CH ₂ CH(CH ₃)CH ₂ -	$-C(CH_3)_2C(=O)CH_3$	resin
1.38	-CH ₂ CH ₂ CH(CH ₃)CH ₂ -	naphth-2-yl	170-171°
1.39	-CH ₂ CH ₂ CH(CH ₃)CH ₂ -	-N(CH ₃)CH ₂ CH ₂ CN	105-106°
1.40	C ₂ H ₅ CH ₃	naphth-2-yl	139-142°

Comp No.	p. R ₂ R ₃	A	Phys.data
1.41	-CH ₂ CH ₂ CHCH ₂ - (CH ₂) ₃ CH(CH ₃) ₂	naphth-2-yl	
1.42	-CH ₂ CH ₂ CHCH ₂ - (CH ₂) ₃ CH(CH ₃) ₂	-C(CH ₃) ₂ OC(=O)CH ₃	resin
1.43 1.44 1.45 1.46 1.47 1.48 1.49 1.50 1.51 1.52 1.53	CH ₃ C ₂ H ₅ CH ₃ C ₂ H ₅ -(CH ₂) ₃ (CH ₂) ₃ (CH ₂) ₃ CH(CH ₃)-(CH ₂) ₃ CH(CH ₃)-(CH ₂) ₃ CH(CH ₃)-(CH ₂) ₃ CH ₂ (CH(CH ₃)) ₂ CH ₂ CH ₂ (CH(CH ₃)) ₂ CH ₂ CH ₂ (CH(CH ₃)) ₂ CH ₂ (CH ₂) ₄ CH ₂ CHCH ₂ CH ₂ - (CH ₂) ₃ CH(CH ₃) ₂	-C(CH ₃) ₂ OC(=O)CH ₃ -N(CH ₃)CH ₂ CH ₂ CN -N(CH ₃)CH ₂ CH ₂ CN naphth-2-yl -C(CH ₃) ₂ OC(=O)CH ₃ -N(CH ₃)CH ₂ CH ₂ CN naphth-2-yl naphth-2-yl -C(CH ₃) ₂ OC(=O)CH ₃ -N(CH ₃)CH ₂ CCN cyclopentyloxy -N(CH ₃)CH ₂ CH ₂ CN	resin resin 82-83° resin resin amorphous 186-187° resin amorphous
1.55 1.56 1.57 1.58 1.59 1.60 1.61 1.62	-(CH ₂) ₄ - CH ₃ CH ₃ CH ₃ CH ₃ -(CH ₂) ₄ (CH ₂) ₄ CH ₂ (CH(CH ₃)) ₂ CH ₂ (CH ₂) ₄ (CH ₂) ₄ (CH ₂) ₄ -	-N(C ₂ H ₅) — -N(CH ₃)CH ₂ CH ₂ CN -N(C ₂ H ₅)CH ₂ CH ₂ CN -N(C ₃ H ₇ -n)CH ₂ CH ₂ CN -N(C ₂ H ₅)CH ₂ CH ₂ CN -N(C ₂ H ₅)CH ₂ CH ₂ CN -N(CH ₂ CH ₂ CN) ₂ -N(CH ₂ CH ₂ CN)C ₄ H ₉ (n) -N(CH ₂ CH ₂ CN)CH ₂	resin 83-85° 112-113° 109-110° 106-107° 122-123° 173-175° 87-89°

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Comp.	R ₂ R ₃	A	Phys.data
1.64	-(CH ₂) ₄ -	-N(CH ₂ CH ₂ CN)	148-150°
1.65	-(CH ₂) ₄ -	-N(CH ₂ CH ₂ CN)CH ₂ CH ₂ OCH ₃	
1.66	-(CH ₂) ₄ -	-N(CH ₂ CH ₂ CN)	111-113°
1.67	-(CH ₂) ₄ -	-N(CH ₂ CH ₂ CN)	126-128°
1.68	-(CH ₂) ₄ -	-N(CH ₂ CH ₂ CN)CH ₂	138-140°
1.69	-(CH ₂) ₄ -	-N(CH2CH2CN)C6H13(n)	79-81°
1.70	-(CH ₂) ₅ -	-N(CH ₃)CH ₂ CH ₂ CN	
1.71	-(CH ₂) ₅ -	-N(C ₂ H ₅)CH ₂ CH ₂ CN	
1.72	-(CH ₂) ₅ -	-N(CH ₂ CH ₂ CN)	
1.73	-(CH ₂) ₅ -	-N(CH ₂ CH ₂ CN)C ₆ H ₁₃ (n)	,
1.74	-(CH ₂) ₅ -	-N(CH₂CH₂CN)	
1.75	-CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ -	-N(CH ₃)CH ₂ CH ₂ CN	
1.76	-CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ -	-N(CH2CH2CN)C6H13(n)	
1.77	-CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ -	-N(CH ₂ CH ₂ CN)	
1.78	-CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ -	-N(C ₂ H ₅)CH ₂ CH ₂ CN	
1.79	-(CH ₂) ₅ -	naphth-2-yl	
1.80	-(CH ₂) ₅ -	-C(CH ₃) ₂	
1.81	-CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ -	naphth-2-yl	
1.82	-CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ -	-C(CH ₃) ₂	
1.83 1.84	-CH ₂ CH(CH ₃)CH ₂ CH ₂ - -CH ₂ CH(CH ₃)CH ₂ CH ₂ -	-N(C ₂ H ₅)CH ₂ CH ₂ CN -N(C ₃ H ₇ -n)CH ₂ CH ₂ CN	

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Comp No.	. R ₂ R ₃	A	Phys.data
1.85	-CH ₂ CH(CH ₃)CH ₂ CH ₂ -	-N(CH ₂ CH ₂ CN)C ₄ H ₉ (n)	
1.86	-CH ₂ CH(CH ₃)CH ₂ CH ₂ -	$-N(CH_2CH_2CN)C_6H_{13}(n)$	
1.87	-CH ₂ CH(CH ₃)CH ₂ CH ₂ -	-N(CH ₂ CH ₂ CN)C ₈ H ₁₇ -n	
1.88	-CH ₂ CH(CH ₃)CH ₂ CH ₂ -	-N(CH ₂ CH ₂ CN)	
1.89	-CH ₂ CH(CH ₃)CH ₂ CH ₂ -	-N(CH ₂ CH ₂ CN)	
1.90	-CH ₂ CH(CH ₃)CH ₂ CH ₂ -	-N(CH ₂ CH ₂ CN)CH ₂	
1.91	-CH ₂ CH(CH ₃)CH ₂ CH ₂ -	-N(CH ₂ CH ₂ CN)	
1.92	-CH ₂ CH(CH ₃)CH ₂ CH ₂ -	-N(CH ₂ CH ₂ CN) ₂	
1.93	-CH ₂ CH(CH ₃)CH ₂ CH ₂ -	-N(CH ₂ CH ₂ CN)CH ₂ CH ₂ OCH ₃	
1.94	-(CH ₂) ₃ CH(CH ₃)-	-N(C ₂ H ₅)CH ₂ CH ₂ CN	
1.95	-(CH ₂) ₃ CH(CH ₃)-	-N(C ₃ H ₇ -n)CH ₂ CH ₂ CN	,
1.96	-(CH ₂) ₃ CH(CH ₃)-	$-N(CH_2CH_2CN)C_4H_9(n)$	
1.97	-(CH ₂) ₃ CH(CH ₃)-	-N(CH2CH2CN)C6H13(n)	
1.98	-(CH ₂) ₃ CH(CH ₃)-	-N(CH ₂ CH ₂ CN)	
1.99	-(CH ₂) ₃ CH(CH ₃)-	-N(CH ₂ CH ₂ CN)CH ₂	
1.100	-(CH ₂) ₅ -	-N(C ₃ H ₇ -n)CH ₂ CH ₂ CN	
1.101	-(CH ₂) ₅ -	$-N(CH_2CH_2CN)C_4H_9(n)$	
1.102	-(CH ₂) ₅ -	-N(CH ₂ CH ₂ CN)C ₆ H ₁₃ (n)	
1.103	-(CH ₂) ₅ -	-N(CH ₂ CH ₂ CN)C ₈ H ₁₇ -n	
.104	-(CH ₂) ₅ -	-N(CH ₂ CH ₂ CN)	

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Comp.	R ₂ R ₃	A	Phys.data
1.105	-(CH ₂) ₅ -	-N(CH ₂ CH ₂ CN)	
-1:406···	-(CH ₂) ₅ -	-N(GHZGHZCN)GHZ	ang sa was
1.107	-(CH ₂) ₅ -	-N(CH ₂ CH ₂ CN)CH ₂	
1.108	-(CH ₂) ₅ -	-N(CH ₂ CH ₂ CN)	
1.109	-(CH ₂) ₅ -	-N(CH ₂ CH ₂ CN) ₂	
1.110	-(CH ₂) ₅ -	-N(CH ₂ CH ₂ CN)CH ₂ CH ₂ OCH ₃	
1.111	-CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ -	-N(C ₃ H ₇ -n)CH ₂ CH ₂ CN	
1.112	-CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ -	-N(CH ₂ CH ₂ CN)C ₄ H ₉ (n)	
1.113	-CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ -	-N(CH ₂ CH ₂ CN)CH ₂	
1.114	-CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ -	-N(CH ₂ CH ₂ CN)CH ₂	•
1.115	-CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ -	-N(CH ₂ CH ₂ CN)	
1.116	-CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ -	-N(CH ₂ CH ₂ CN) ₂	
1.117	-CH ₂ CH(CH ₃)CH ₂ CH ₂ -	-N(CH ₂ CH ₂ CN)CH ₂	
1.118	CH ₃ CH ₃	-N(CH ₂ CH ₂ CN)	172-174°
1.119	-CH ₂ (CH(CH ₃)) ₂ CH ₂ -	-N(CH ₂ CH ₂ CN)CH ₂	155-157°
1.120	-CH2(CH(CH3))2CH2-	-N(CH2CH2CN)C4H9(n)	104-106°
1.121	-(CH ₂) ₄ -	-N(CH ₂ CH ₂ CN)	164-166°

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Comp.	R ₂ R ₃	A	Phys.data
1.122	-(CH ₂) ₄ -	-N(CH ₂ CH ₂ CN)C ₈ H ₁₅ -c	
1.123	-(CH ₂) ₄ -	-N(CH ₂ CH ₂ CN)C ₇ H ₁₃ -c	
1.124	-(CH ₂) ₄ -	-N(CH ₂ CH ₂ CN)C ₁₂ H ₂₃ -c	154-156°
1.125	-(CH ₂) ₄ -	-N(CH ₂ CH ₂ CN)-(CH ₂) ₃ OCH ₃	

Table 2

Comp. No.	R ₂	R ₃	A	R ₄	Phys.data
2.1	-(CH ₂) ₄	-	-CH ₂ —	6-Cl	142-143°
2.2	-(CH ₂) ₄	.	-CH ₂ CH ₂	6-Cl	106-108°
2.3	-(CH ₂) ₄	-	-CH ₂ —	6-F	resin
2.4	-(CH ₂) ₄ ·	-	-CH ₂	6-F	resin
2.5	-(CH ₂) ₄ -	-	-CH ₂ CH ₂	6-F	resin
2.6	-(CH ₂) ₄ -	-		6-C1	208°
2.7	-(CH ₂) ₄ -			6-Cl	220°
2.8	-(CH ₂) ₄ -		-CH ₂	6-Cl	136-139°

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Comp.	R ₂	R ₃	A	R ₄	Phys.data
2.9	-(CH ₂) ₄	-	-CH ₂ —	6-Cl	106-109°
2.10	-(CH ₂) ₄	-	-CH ₂ CH ₂	6-Cl	97-100°
2.11	-(CH ₂) ₄	-	-C(CH ₃) ₂ OCOCH ₃	6-Cl	113-116°
2.12	-(CH ₂) ₄	-	-CH(C ₆ H ₅)-OCOCH ₃	6-Cl	156-158°
2.13	-(CH ₂) ₄	-	-CH ₂ OCH ₂ C ₆ H ₅	6-Cl	230°
2.14	-(CH ₂) ₄	-	-C(CH ₃) ₂ —	6-C1	117-119°
2.15	-(CH ₂) ₄ ·	-	-C(CH ₃) ₂ -OCO-t-C ₄ H ₉	6-C1	151-153°
2.16	-(CH ₂) ₄ ·	-	-C(CH ₃) ₂ -OCO-C ₆ H ₅	6-Cl	amorphous
2.17	-(CH ₂) ₄ -	-	-N(CH ₃)CH ₂ CH ₂ CN	6-Cl	156-158°
2.18	$-C_2H_5$	-C ₂ H ₅	-C(CH ₃) ₂ -OCO-t-C ₄ H ₉	6-Cl	90-91°
2.19	-C ₂ H ₅	-C ₂ H ₅	-C(CH ₃) ₂	6-Cl	141-142°
2.20	-(CH ₂) ₄ -	•	-N(C ₂ H ₅)CH ₂ CH ₂ CN	6-C1	147-148°
2.21	-(CH ₂) ₄ -		-N(C ₃ H ₇ -n)CH ₂ CH ₂ CN	6-Cl	134-135°
2.22	-(CH ₂) ₄ -		$-N(CH_2CH_2CN)C_6H_{13}(n)$	6-Cl	78-80°
2.23	-(CH ₂) ₄ -		-N(CH ₂ CH ₂ CN)CH ₂	6-Cl	159-161°
2.24	-(CH ₂) ₄ -		-N(CH ₂ CH ₂ CN)C ₄ H ₉ (n)	6-Cl	127-129°
2.25	-(CH ₂) ₄ -		-N(CH ₂ CH ₂ CN)	6-Cl	223-225°
2.26	-(CH ₂) ₄ -		-N(CH ₂ CH ₂ CN)C ₈ H ₁₇ -n	6-Cl	
2.27	-(CH ₂) ₄ -		-N(C ₃ H ₇ -n)CH ₂ CH ₂ CN	6-F	
2.28	-(CH ₂) ₄ -		-N(CH ₂ CH ₂ CN) ₂	6-Cl	188-190°
2.29	-(CH ₂) ₄ -		-N(CH ₂ CH ₂ CN)	6-C1	125-127°

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Con No.	np. R ₂	R ₃	A	R ₄	Phys.data
2.30	-(CH ₂)4-	-N(CH2CH2CN)	6-CI	155-157°
2.31	-(CH ₂)3-	-N(CH ₃)CH ₂ CH ₂ CN	6-Cl	
2.32	-(CH ₂)3-	-N(CH ₃)CH ₂ CH ₂ CN	6-F	
2.33	-(CH ₂))3-	-N(CH ₂ CH ₂ CN)CH ₂	6-Cl	
2.34	-(CH ₂))3-	-N(CH ₂ CH ₂ CN)CH ₂		
2.35	-(CH ₂))3-	$-N(CH_2CH_2CN)C_6H_{13}(n)$	6-C1	
2.36	-(CH ₂)) ₃ -	$-N(CH_2CH_2CN)C_6H_{13}(n)$	6-F	
2.37	-CH ₂ CH	I(CH ₃)CH ₂ CH ₂ -		6-CI	
2.38	-CH ₂ CH	I(CH ₃)CH ₂ CH ₂ -	-N(C ₂ H ₅)CH ₂ CH ₂ CN	6-C1	
2.39	-CH ₂ CH	I(CH ₃)CH ₂ CH ₂ -	-N(C ₃ H ₇ -n)CH ₂ CH ₂ CN	6-Cl	
2.40	-CH ₂ CH	(CH ₃)CH ₂ CH ₂ -		6-Cl	
2.41	-CH ₂ CH	(CH ₃)CH ₂ CH ₂ -		6-Cl	
2.42	-CH ₂ CH	(CH ₃)CH ₂ CH ₂ -		6-Cl	
2.43	-CH ₂ CH	(CH ₃)CH ₂ CH ₂ -	-N(CH ₂ CH ₂ CN)	6-C1	
2.44	-CH ₂ CH((CH ₃)CH ₂ CH ₂ -	-N(CH₂CH₂CN)CH₂ _	6-Cl	
2.45	-CH ₂ CH((CH ₃)CH ₂ CH ₂ -	-N(CH ₂ CH ₂ CN)	6-Cl	
2.46	-CH ₂ CH(CH ₃)CH ₂ CH ₂ -	-N(CH ₂ CH ₂ CN) ₂	6-Cl	
2.47		CH ₃)CH ₂ CH ₂ -		CH ₃ 6-Cl	
2.48		CH(CH ₃)-	-N(CH ₃)CH ₂ CH ₂ CN	6-C1	
2.49	-(CH ₂) ₃ (CH(CH ₃)-	-N(C ₂ H ₅)CH ₂ CH ₂ CN	6-Cl	
2.50		CH(CH ₃)-	-N(C ₃ H ₇ -n)CH ₂ CH ₂ CN	6-Cl	
2.51		CH(CH ₃)-	$-N(CH_2CH_2CN)C_4H_9(n)$	6-Cl	

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Comp No.	. R ₂	R ₃	A	R ₄ Phys. data
2.52	-(CH ₂)) ₃ CH(CH ₃)-	-N(CH ₂ CH ₂ CN)C ₆ H ₁₃ (n)	6-Cl
2.53	-(CH ₂)) ₃ CH(CH ₃)-	-N(CH ₂ CH ₂ CN)	6-Cl
2.54	-(CH ₂)) ₃ CH(CH ₃)-	-N(CH ₂ CH ₂ CN)	6-C1
2.55	-(CH ₂)	₃ CH(CH ₃)-	-N(CH2CH2CN)CH2	6-Cl
2.56	-(CH ₂)	₃ CH(CH ₃)-	-N(CH ₂ CH ₂ CN)	6-Cl
2.57	-(CH ₂) ₃	₃ CH(CH ₃)-	-N(CH ₂ CH ₂ CN) ₂	6-C1
2.58	-(CH ₂) ₃	3CH(CH ₃)-	-N(CH ₂ CH ₂ CN)CH ₂ CH ₂ OCH ₃	6-Cl
2.59	-(CH ₂) ₅	5-	-N(CH ₃)CH ₂ CH ₂ CN	6-C1
2.60	-(CH ₂) ₅	5-	-N(C ₂ H ₅)CH ₂ CH ₂ CN	6-Cl
2.61	-(CH ₂) ₅	5-	-N(C ₃ H ₇ -n)CH ₂ CH ₂ CN	6-Cl
2.62	-(CH ₂) ₅	5-	-N(CH ₂ CH ₂ CN)C ₄ H ₉ (n)	6-Cl
2.63	-(CH ₂) ₅	5-	-N(CH ₂ CH ₂ CN)C ₆ H ₁₃ (n)	6-C1
2.64	-(CH ₂) ₅	. -	-N(CH ₂ CH ₂ CN)	6-Cl
2.65	-(CH ₂) ₅	.	-N(CH ₂ CH ₂ CN)	6-Cl
2.66	-(CH ₂) ₅ .	-	-N(CH ₂ CH ₂ CN)CH ₂	6-Cl
2.67	-(CH ₂) ₅ -	-	-N(CH₂CH₂CN)	6-Cl
2.68	-(CH ₂) ₅ -		-N(CH ₂ CH ₂ CN) ₂	6-Cl
2.69	-(CH ₂) ₅ -		-N(CH ₂ CH ₂ CN)CH ₂ CH ₂ OCH ₃	6-Ci
2.70	-CH ₂ CH	I ₂ CH(CH ₃)CH ₂ CH ₂ -		6-Cl
2.71		2CH(CH ₃)CH ₂ CH ₂ -		6-Cl

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Comp.	R ₂ R ₃		A	R ₄ Phys.
2.72	-CH ₂ CH ₂ CH(CI		-N(C ₃ H ₇ -n)CH ₂ CH ₂ CN	6-Cl
2.73	-CH ₂ CH ₂ CH(CH		-N(CH ₂ CH ₂ CN)C ₄ H ₉ (n)	6-Cl
2.74	-CH ₂ CH ₂ CH(CH	H ₃)CH ₂ CH ₂ -	-N(CH2CH2CN)C6H13(n)	6-C1
2.75	-CH ₂ CH ₂ CH(CI	H ₃)CH ₂ CH ₂ -	-N(CH ₂ CH ₂ CN)	6-Cl
2.76	-CH ₂ CH ₂ CH(CF	H ₃)CH ₂ CH ₂ -	-N(CH ₂ CH ₂ CN)	6-C1
2.77	-CH ₂ CH ₂ CH(CH	H ₃)CH ₂ CH ₂ -	-N(CH ₂ CH ₂ CN)CH ₂	6-Cl
2.78	-CH ₂ CH ₂ CH(CF	H ₃)CH ₂ CH ₂ -	-N(CH ₂ CH ₂ CN)	6-Cl
2.79	-CH ₂ CH ₂ CH(CH	H ₃)CH ₂ CH ₂ -	-N(CH ₂ CH ₂ CN) ₂	6-Cl
2.80	-CH ₂ CH ₂ CH(CH	H ₃)CH ₂ CH ₂ -	-N(CH ₂ CH ₂ CN)CH ₂ CH ₂ OCH ₃	6-Cl
2.81	-CH ₂ CH ₂ CH(CH	H ₃)CH ₂ CH ₂ -	naphth-2-yl	6-Cl
2.82	-(CH ₂) ₅ -		naphth-2-yl	6-Cl
2.83	-CH ₂ CH(CH ₃)C	H ₂ CH ₂ -	naphth-2-yl	6-C1
2.84	-(CH ₂) ₃ CH(CH ₃)-	naphth-2-yl	6-Cl
2.85	CH ₃ CH ₃		naphth-2-yl	6-C1
2.86	-(CH ₂) ₄ -		-N(CH ₃)CH ₂ CH ₂ CN	6-F
2.87	-(CH ₂) ₄ -		-N(C ₂ H ₅)CH ₂ CH ₂ CN	6-F
2.88	-(CH ₂) ₄ -		$-N(CH_2CH_2CN)C_6H_{13}(n)$	6-CH ₃
2.89	-(CH ₂) ₄ -		-N(CH ₂ CH ₂ CN)C ₄ H ₉ (n)	6-F
2.90	-(CH ₂) ₄ -		-N(CH2CH2CN)C6H13(n)	6-F
2.91	-(CH ₂) ₄ -		-N(CH ₂ CH ₂ CN)	6-F
2.92	-(CH ₂) ₄ -		-N(CH ₂ CH ₂ CN)	6-F
2.93	-(CH ₂) ₄ -		-N(CH ₂ CH ₂ CN)CH ₂	6-F

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Comp.	R ₂	R ₃	A	R ₄ Phys. data
2.94	-(CH ₂) ₄ :	-	-N(CH ₂ CH ₂ CN)	6-F
2.95	-(CH ₂) ₄ -		-N(CH ₂ CH ₂ CN) ₂	6-F
2.96	-(CH ₂) ₄ -		-N(CH ₂ CH ₂ CN)CH ₂ CH ₂ OCH ₃	6-F
2.97	-(CH ₂) ₄ -		naphth-2-yl	6- F
2.98	-(CH ₂) ₄ -		-N(CH ₃)CH ₂ CH ₂ CN	6-CH ₃
2.99	-(CH ₂) ₄ -		-N(C ₂ H ₅)CH ₂ CH ₂ CN	6-CH ₃
2.100	-(CH ₂) ₄ -		-N(C ₃ H ₇ -n)CH ₂ CH ₂ CN	6-CH ₃
2.101	-(CH ₂) ₄ -		-N(CH2CH2CN)C4H9(n)	6-CH ₃
2.102	-(CH ₂) ₄ -		-N(CH ₂ CH ₂ CN)CH ₂	6-Cl
2.103	-(CH ₂) ₄ -		-N(CH ₂ CH ₂ CN)CH ₂	6-CH ₃

Table 3

$$\begin{array}{c|c}
 & O \\
 & \parallel \\
 & R_3 \\
 & N \\
 & R_2
\end{array}$$

$$\begin{array}{c}
 & O \\
 & \parallel \\
 & O \\
 & C \\
 & -A \\
 & (R_4)_n
\end{array}$$
(Ic)

Cpd. No.	R ₂ R ₃	A	R ₄	n Phys.data
3.1	-(CH ₂) ₄ -	-CH ₂ -	2-CH ₃ ,6-CH ₃	2 77-79°
3.2	CH ₃ CH ₃		2-CH ₃	1 98-100°
3.3	CH ₃ CH ₃	-C(CH ₃) ₂ -OCOCH ₃	2-CH ₃	1 118-120°
3.4	-(CH ₂) ₄ -	ларhth-2-yl	2-CH ₃ ,3-CH ₃ ,	
			6-CH ₃	3
3.5	$-(CH_2)_4$ -	-N(CH ₃)CH ₂ CH ₂ CN	2-CH ₃ ,6-CH ₃	2 .
3.6	-(CH ₂) ₅ -	-N(CH ₃)CH ₂ CH ₂ CN	2-CH ₃ ,6-CH ₃	2
3.7	-(CH ₂) ₃ -	-N(CH ₃)CH ₂ CH ₂ CN	2-CH ₃ ,6-CH ₃	2
3.8	-(CH ₂) ₄ -	naphth-2-yl	2-CH ₃ ,6-CH ₃	2
3.9	-(CH ₂) ₄ -	naphth-2-yl	2-CH ₃ ,6-C ₂ H ₅	2
3.10	-(CH ₂) ₅ -	naphth-2-yl	2-CH ₃ ,6-CH ₃	2
3.11	-(CH ₂) ₃ -	naphth-2-yl	2-CH ₃ ,6-CH ₃	2

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Cpd.	. R ₂ R ₃	Α	R ₄	n	Phys. data
3.12	-CH ₂ CH(CH ₃)CH ₂ CH ₂ -	naphth-2-yl	2-CH ₃ ,		
			6-CH ₃	2	
3.13	-CH(CH ₃)-(CH ₂) ₃ -	naphth-2-yl	2-CH ₃ ,		
			6-CH ₃	2	
3.14	-CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ -	naphth-2-yl	2-CH ₃ ,		
			6-CH ₃	2	
3.15	-(CH ₂) ₄ -	-N(CH2CH2CN)C6H13(n)	2-CH ₃ ,		
			6-CH ₃	2	
3.16	-CH ₂ CH(CH ₃)CH ₂ CH ₂ -	-N(CH ₃)CH ₂ CH ₂ CN	2-CH ₃ ,		
2 12			6 -CH $_3$	2	
3.17	-CH(CH ₃)-(CH ₂) ₃ -	-N(CH ₃)CH ₂ CH ₂ CN	$2-CH_3$,		
3.18			$6-CH_3$	2	
3.18	-CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ -	-N(CH ₃)CH ₂ CH ₂ CN	$2-CH_3$,		
3.19	-(CH ₂) ₄ -	N/CH \CH CH CH	6-CH ₃	2	
3.19	-(CH ₂) ₄ -	-N(CH ₃)CH ₂ CH ₂ CN	2-CH ₃ ,		
			3-CH ₃ ,		
3.20	-(CH ₂) ₃ -	N/CU \CH CH CH	6-CH ₃	3	
2,20	(0.12)3-	-N(CH ₃)CH ₂ CH ₂ CN	2-CH ₃ ,		
			3-CH ₃ ,	2	
3.21	-(CH ₂) ₅ -	-N(CH ₃)CH ₂ CH ₂ CN	6-CH₃ 2-CH₃,	3	
	2.3	14(613)61126112614	2-CH ₃ , 3-CH ₃ ,		
			6-CH ₃	3	
3.22	-CH ₂ CH(CH ₃)CH ₂ CH ₂ -	-N(CH ₃)CH ₂ CH ₂ CN	2-CH ₃ ,	5	
	2 3 2 2		3-CH ₃ ,		
			6-CH ₃	3	
3.23	-CH ₂ CH ₂ CH(CH ₃)CH ₂ CH ₂ -	-N(CH₃)CH₂CH₂CN	2-CH ₃ ,	5	
			3-CH ₃ ,		
			6-CH ₃	3	

Table 4

$$\begin{array}{c|c}
C & C & A \\
R_3 & C_2H_5 & C_2H_5 \\
R_2 & C_3H_5
\end{array}$$
(Id)

Comp.No.	R ₂ R ₃	Α	Phys.data
4.1	-(CH ₂) ₄ -	-CH ₂ —	
4.2	-(CH ₂) ₄ -	-CH ₂	
4.3	-(CH ₂) ₄ -	-CH(C ₆ H ₅)OCOCH ₃	
4.4	-(CH ₂) ₄ -	-CH(C ₆ H ₅)OCO-i-C ₃ H ₇	
4.5	-(CH ₂) ₄ -	-CH(C ₆ H ₅)OCO-t-C ₄ H ₉	
4.6	-(CH ₂) ₄ -	-C(CH ₃) ₂ OCOCH ₃	
4.7	-(CH ₂) ₄ -	-C(CH ₃) ₂ OCO-i-C ₃ H ₇	•
4.8	-(CH ₂) ₄ -	-C(CH ₃) ₂ OCO-t-C ₄ H ₉	
4.9	-(CH ₂) ₄ -	-C(CH ₃) ₂ OCOC ₆ H ₅	
4.10	-(CH ₂) ₄ -		
4.11	-(CH ₂) ₄ -		
4.12	-(CH ₂) ₄ -	-C(CH ₃) ₂ —	119-120°

Comp.l	No. R ₂ R ₃	Α	Phys.data
4.13	(CH.)		
4.13	-(CH ₂) ₄ -	CH ₃	
4.14	-(CH ₂) ₄ -	∑ CH₃	
4.15	-(CH ₂) ₄ -		132-134°
		×C ₆ H ₅	
4.16	-(CH ₂) ₄ -	\triangle	oil
4.17	CH ₃ CH ₃	-CH(C ₆ H ₅)OCOCH ₃	•
4.18	CH ₃ CH ₃	-C(CH ₃) ₂ OCOCH ₃	
4.19	CH ₃ CH ₃	-CH ₂ —	
		CH₃	
4.20	CH ₃ CH ₃	Δ	
4.21	CH ₃ CH ₃	-C(CH ₃) ₂ —	82-83°
4.22	-(CH ₂) ₄ -	cyclohexyloxy	oil
4.23	-(CH ₂) ₅ -	cyclohexyloxy	
4.24	-CH(CH ₃)-(CH ₂) ₄ -	cyclohexyloxy	
4.25	-(CH ₂) ₃ -	cyclohexyloxy	

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Table 5

$$R_3$$
 N
 CH_3
 CH_3
 CH_3
 CH_3
 CH_3

Comj No.	p. R ₂ R ₃	B	Phys. data
5.1	-(CH ₂) ₄ -	-NH(c-C ₃ H ₅)	
5.2	-(CH ₂) ₄ -	$-NH(c-C_6H_{11})$	
5.3	-(CH ₂) ₄ -	-NH(c-C ₈ H ₁₅)	
5.4	-(CH ₂) ₄ -	-N(CH2-c-C3H5)2	
5.5	-(CH ₂) ₄ -	$-N(c-C_3H_5)_2$	
5.6	-(CH ₂) ₄ -	$-N(c-C_6H_{11})_2$	
5.7	-(CH ₂) ₄ -	-N(CH3)(c-C3H5)	
5.8	-(CH ₂) ₄ -	$-N(CH_3)(c-C_6H_{11})$	171-1 7 3°
5.9	-(CH ₂) ₄ -	$-N(C_2H_5)(c-C_6H_{11})$	
5.10	-(CH ₂) ₄ -	$-N(n-C_3H_7)(c-C_6H_{11})$	
5.11	-(CH ₂) ₄ -	$-N(n-C_3H_7)(CH_2-c-C_3H_5)$,
5.12	-(CH ₂) ₄ -	-N(CH ₃)(CH ₂ CH ₂ OCH ₃)	
5.13	-(CH ₂) ₄ -	$-N(C_2H_5)(CH_2CH_2OCH_3)$	
5.14	-(CH ₂) ₄ -	$-N(n-C_3H_7)(CH_2CH_2OCH_3)$	
5.15	-(CH ₂) ₄ -	-N(CH ₂ CH ₂ OCH ₃) ₂	resin
5.16	-(CH ₂) ₄ -	-N(CH ₂ CH ₂ SCH ₃) ₂	
5.17	-(CH ₂) ₄ -	-N(CH2CH2OC2H5)2	
5.18	-(CH ₂) ₄ -	-N(CH ₂ CH ₂ OCH ₂ CH ₂ OCH ₃) ₂	
5.19	-(CH ₂) ₄ -	-N(CH ₂ CH ₂ OCH ₂ CH ₂ OC ₂ H ₅) ₂	
5.20	-(CH ₂) ₄ -	$-N(CH_2CH=CH_2)_2$	
5.21	-(CH ₂) ₄ -	$-N(CH_3)(CH_2CH=CH_2)$	
5.22	-(CH ₂) ₄ -	$-N(C_2H_5)(CH_2CH=CH_2)$	
5.23	-(CH ₂) ₄ -	$-N(n-C_3H_7)(CH_2CH=CH_2)$	
5.24	-(CH ₂) ₄ -	$-N(i-C_3H_7)(CH_2CH=CH_2)$	
5.25	-(CH ₂) ₄ -	$-N(c-C_3H_5)(CH_2CH=CH_2)$	
5.26	-(CH ₂) ₄ -	$-N(c-C_6H_{11})(CH_2CH=CH_2)$	

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Comp.	R ₂ F	В В	Phys. data
5.27	-(CH ₂)	- N(CH ₃)(CH ₂ -C≡CH)	
5.28	-(CH ₂)		
5.29	-(CH ₂)		
5.30	-(CH ₂),		
5.31	-(CH ₂),		
5.32	-(CH ₂)	- $-N(C_2H_5)[CH(CH_3)C\equiv CH]$	
5.33	-(CH ₂)	- $-N(n-C_3H_7)[CH(CH_3)C\equiv CH$	ŋ
5.34	-(CH ₂) ₄	- NH-CH ₂ -C≡CH	
5.35	-(CH ₂) ₄	- NH-CH(CH ₃)-C≡CH	
5.36	-(CH ₂) ₄	- NH-C(CH ₃) ₂ -C≡CH	
5.37	-(CH ₂) ₄	- NH-CH(C ₂ H ₅)-C≡CH	
5.38	-(CH ₂) ₄	- NH-C(C_2H_5) ₂ -C=CH	
5.39	-(CH ₂) ₄	- NH-CH(CH ₃)-CH=CH ₂	
5.40	-(CH ₂) ₄	-NH-C(CH ₃) ₂ -CH=CH ₂	
5.41	-(CH ₂) ₄	$-NH-C(C_2H_5)_2-CH=CH_2$	
5.42	-(CH ₂) ₄	-N(CH ₃)CH ₂ CH ₂ CN	
5.43	-(CH ₂) ₄	-N(C ₂ H ₅)CH ₂ CH ₂ CN	
5.44	$-(CH_2)_4$	-N(n-C ₃ H ₇)CH ₂ CH ₂ CN	•
5.45	-(CH ₂) ₄	-N(CH ₂ CH ₂ CN) ₂	
5.46	-(CH ₂) ₄	$-N(CH_3)(CH_2C_6H_5)$	
5.47	-(CH ₂) ₄	$-N(C_2H_5)(CH_2-C_6H_5)$	resin
5.48	-(CH ₂) ₄ ·	$-N(n-C_3H_7)(CH_2-C_6H_5)$	
5.49	-(CH ₂) ₄ ·	$-N(i-C_3H_7)(CH_2-C_6H_5)$	
5.50	-(CH ₂) ₄ ·	-N(CH2-C6H5)2	
5.51	-(CH ₂) ₄ -	-N(CH3)(CH2-C6H4-4-Cl)	
5.52	-(CH ₂) ₄ -	-N(CH ₃)(CH ₂ -C ₆ H ₃ -3,4-Cl ₂)	
5.53	-(CH ₂) ₄ -	$-N(CH_3)(CH_2-C_6H_4-4-CH_3)$	
5.54	-(CH ₂) ₄ -	$-N(CH_3)(CH_2-C_6H_4-4-CF_3)$	
5.55	$-(CH_2)_4$	$-N(C_2H_5)(CH_2-C_6H_4-4-CI)$	
5.56	-(CH ₂) ₄ -	-NH-C ₆ H ₅	
5.57	-(CH ₂) ₄ -	<u>-</u> •	
5.58	-(CH ₂) ₄ -	-NH-(Naphthyl-2)	
5.59	-(CH ₂) ₄ -	-NH-C ₆ H ₄ -4-Cl	

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Comp.	R ₂ R ₃	В	Phys. data
5.60	-(CH ₂) ₄ -	-NH-C₀H₄-4-CH₃	
5.61	-(CH ₂) ₄ -	-NH-C ₆ H ₄ -4-OCH ₃	
5.62	-(CH ₂) ₄ -	$-NH-C_6H_4-4-NO_2$	
5.63	-(CH ₂) ₄ -	-NH-C ₆ H ₄ -4-CN	يوسيد برياد فالانتهاد برياد
5.64	-(CH ₂) ₄ -	-NH-C ₆ H ₄ -4-CF ₃	
5.65	-(CH ₂) ₄ -	-NH-C ₆ H ₃ -3,4-(CH ₃) ₂	•
5.66	-(CH ₂) ₄ -	-NH-C ₆ H ₃ -3,4-Cl ₂	
5.67	-(CH ₂) ₄ -	-NH-C ₆ H ₃ -3,5-(CF ₃) ₂	
5.68	-(CH ₂) ₄ -	-NH-C ₆ H ₄ -3-CF ₃	
5.69	-(CH ₂) ₄ -	-NH-C ₆ H ₄ -4(O-C ₆ H ₅)	
5.70	-(CH ₂) ₄ -	-NH-C ₆ H ₄ -4(S-C ₆ H ₅)	
5.71	-(CH ₂) ₄ -	-NH-C ₆ H ₄ -2-CH ₃	
5.72	-(CH ₂) ₄ -	-NH-C ₆ H ₄ -2-C ₂ H ₅	
5.73	-(CH ₂) ₄ -	-NH-C ₆ H ₄ -2-(i-C ₃ H ₇)	
5.74	-(CH ₂) ₄ -	-NH-C ₆ H ₃ -2,6-(CH ₃) ₂	
5.75	-(CH ₂) ₄ -	$-NH-C_6H_3-2,6-(C_2H_5)_2$	
5.76	-(CH ₂) ₄ -	-NH-C ₆ H ₃ -2,6-(i-C ₃ H ₇) ₂	
5.77	-(CH ₂) ₄ -	-NH-C ₆ H ₄ -4-F	•
5.78	-(CH ₂) ₄ -	$-N-(C_6H_5)_2$	
5.79	-(CH ₂) ₄ -	-N(CH3)(C6H5)	
5.80	-(CH ₂) ₄ -	$-N(C_2H_5)(C_6H_5)$	166-168°
5.81	-(CH ₂) ₄ -	-N(CH ₃)(C ₆ H ₄ -4-Cl)	
5.82	-(CH ₂) ₄ -	$-N(CH_3)(C_6H_4-4-CH_3)$	
5.83	-(CH ₂) ₄ -	$-N(CH_3)(C_6H_4-4-F)$	
5.84	-(CH ₂) ₄ -	-N(CH ₃)(C ₆ H ₃ -3,4-Cl ₂)	
5.85	-(CH ₂) ₄ -	$-N(CH_3)[C_6H_3-3,4-(CH_3)_2]$	
5.86	-(CH ₂) ₄ -	$-N(CH_3)[C_6H_4-4(O-C_6H_5)]$	
5.87	-(CH ₂) ₄ -	$-N(CH_3)[C_6H_4-4(S-C_6H_5)]$	
5.88	-(CH ₂) ₄ -	morpholin-4-yl	resin
5.89	-(CH ₂) ₄ -	4-Thiomorpholinyl	
5.90	-(CH ₂) ₄ -	2,6-Dimethyl-4-morpholinyl	
5.91	-(CH ₂) ₄ -	2,6-Dimethylthio-4-morpholinyl	
5.92	-(CH ₂) ₄ -	1-Piperidyl	

Comp No.	o. R ₂	R ₃	В	Phys. data
5.93	-(CH ₂) ₄ -	1-Pyrrolidinyl	
5.94	-(CH ₂) ₄ -	4-Methyl-piperazin-1-yl	
5.95	-(CH ₂) ₄ -	4-Phenyl-piperazin-1-yl	
5.96	-((CH ₂) ₄ -	4-(2-Methoxyphenyl)-piperazin-1-yl	
5.97	-(0	CH ₂) ₄ -	4-(4-Fluorophenyl)piperazin-1-yl	
5.98	-((CH ₂) ₄ -	4-(4-Nitrophenyl)piperazin-1-yl	
5.99	-((CH ₂) ₄ -	(4-Ethoxycarbonyl)piperazin-i-yl	
5.100	-((CH ₂) ₄ -	4-Benzylpiperazin-1-yl	
5.101	-((CH ₂) ₄ -	4-(2-Pyridyl)piperazin-1-yl	
5.102	-(0	CH ₂) ₄ -	4-(3,4-Dichlorophenyl)piperazin-1-yl	
5.103	-(C	$CH_2)_4$ -	4-(4-Methoxyphenyl)piperazin-1-yl	
5.104	-(C	CH ₂) ₄ -	4-Ethylpiperazin-1-yl	
5.105		CH ₂) ₄ -	1,2,3,4-Tetrahydroquinolin-1-yl	
5.106	-(C	$(H_2)_4$	1,2,3,4-Tetrahydroisoquinolin-2-yl	
5.107	-(C	CH ₂) ₄ -	Indolin-1-yl	139-141°
5.108	-(C	CH ₂) ₄ -	2-Methyl-1-piperidyl	
5.109	-(C	CH ₂) ₄ -	2,6-Dimethyl-1-piperidyl	•
5.110	-(C	H ₂) ₄ -	3,3-Dimethyl-1-piperidyl	49-51°
5.111	-(C	:H ₂) ₄ -	Naphth-1-yl	185-188°
5.112	-(C	H ₂) ₄ -	Naphth-2-yl	163-165°
5.113		H ₂) ₄ -	Naphth-1-oyl	
5.114		H ₂) ₄ -	Naphth-2-oyl	
5.115		H ₂) ₄ -	$-NH(c-C_5H_9)$	
5.116	-(C	H ₂) ₄ -	-NH(c-C ₇ H ₁₃)	
5.117	-(C	H ₂) ₄ -	$-NH(c-C_8H_{15})$	
5.118	-(C)	H ₂) ₄ -	$-NH(c-C_3H_5)$	
5.119	-(C)	H ₂) ₄ -	-NH-Adamantyl-1	
5.120	-(CI	H ₂) ₄ -	$-NH-C_6H_4-(4-c-C_6H_{11})$	
5.121	-(CI	H ₂) ₄ -	$NH-C_6H_4-(4-C_6H_5)$	
5.122	-CH ₃	-CH ₃	1-Naphthyl	184-185°
5.123	-CH ₃	-CH ₃	2-Naphthyl	
5.124	-CH ₃	-CH ₃	$-N(CH_3)-c-C_6H_{11}$	
5.125	-CH ₃	-CH ₃	-N(CH ₂ CH ₂ OCH ₃) ₂	

Comp.	R ₂	R ₃	В	Phys. data
5.126	-CH ₃	-CH ₃	N(CH ₂ CH ₃)C ₆ H ₅	
5.127	-CH ₃	-CH ₃	— N	week w
5.128	-CH ₃	-CH ₃	$-N$ CH_3	
5.129	-CH ₃	-CH ₃	-N_O	
5.130	-CH ₃	$-C_2H_5$	1-Naphthyl	
			2-Naphthyl	
5.132			-N(CH ₃)-c-C ₆ H ₁₁	
5.133		_	-N(CH ₂ CH ₂ OCH ₃) ₂	
5.134	-CH ₃	$-C_2H_5$	N(CH ₂ CH ₃)C ₆ H ₅	
5.135	-CH ₃	-C ₂ H ₅		
5.136	-CH ₃	-C ₂ H ₅	—N ← CH ₃	
5.137	-CH ₃	-C ₂ H ₅	- N_O	
5.138	-C ₂ H ₅	$-C_2H_5$	1-Naphthyl	
5.139	$-C_2H_5$	$-C_2H_5$	2-Naphthyl	
5.140	$-C_2H_5$	$-C_2H_5$	$-N(CH_3)-c-C_6H_{11}$	
5.141	$-C_2H_5$	$-C_2H_5$	-N(CH ₂ CH ₂ OCH ₃) ₂	
5.142	$-C_2H_5$	$-C_2H_5$	$N(CH_2CH_3)C_6H_5$	

Phys. data

Comp.	R ₂	R ₃	В
5.143	-C ₂ H ₅	-C ₂ H ₅	-N
5.144	-C ₂ H ₅	-C ₂ H ₅	$-N$ CH_3
5.145	-C ₂ H ₅	-C ₂ H ₅	-N_O
5.146	-СН ₂ -СН-0 СН ₃ 0		1-Naphthyl
5.147	-СН ₂ -СН-С	СН-СН ₂ - СН ₃	2-Naphthyl
5.148	-сн ₂ -сн-с Сн ₃ с		-N(CH ₃)-c-C ₆ H ₁₁
5.149	-сн ₂ -сн-с 1 сн ₃ с		-N(CH ₂ CH ₂ OCH ₃) ₂
5.150	-сн ₂ -сн-с сн ₃ с		N(CH ₂ CH ₃)C ₆ H ₅
5.151	-сн ₂ -сн-с сн ₃ с	_	-N
5.152	-сн ₂ -сн-с сн ₃ с	_	$-N$ CH_3
5.153	-СН ₂ -СН-СІ СН ₃ С	_	-N_O
5.154	-(CH ₂)3-	1-Naphthyl

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Con No.	np. R ₂ R ₃	В	Phys. data
5.15	5 -(CH ₂) ₃ -	2-Naphthyl	-
5.15	6 -(CH_2) ₃ -	$-N(CH_3)-c-C_6H_{11}$	
5.15	7 $-(CH_2)_3$ -	-N(CH ₂ CH ₂ OCH ₃) ₂	
5.15	8 $-(CH_2)_3$	N(CH ₂ CH ₃)C ₆ H ₅	والمعافلات والمعادية
5.15	9 -(CH ₂) ₃ -	-N	
5.16	0 -(CH ₂) ₃ -	$-N$ CH_3	
5.16	1 -(CH ₂) ₃ -	-N_O	
5.16	2 -(CH ₂) ₄ -	2-Ethyl-1-piperidyl	
5.16		3-Methyl-1-piperidyl	
5.16	$-(CH_2)_4$	4-Methyl-1-piperidyl	125-127°

OSO₂-B

Table 6

Comp. No.	R ₂	R ₃	В	R ₄	Phys. data
6.15	-(CH ₂)) ₃ -	$-N$ CH_3	6-Cl	
6.16	-(CH ₂)	3 -	-N_O	6-Cl	and the second s
6.17	-CH ₃	-CH ₃	1-Naphthyl	6-Cl	
6.18	-CH ₃	-CH ₃	2-Naphthyl	6-C1	
6.19	$-CH_3$	-CH ₃	$-N(CH_3)-c-C_6H_{11}$	6-Cl	
6.20	-CH ₃	-CH ₃	-N(CH ₂ CH ₂ OCH ₃) ₂	6-C1	
6.21	-CH ₃	-CH ₃	N(CH ₂ CH ₃)C ₆ H ₅	6-C1	
6.22	-CH ₃	-CH₃	-N	6-Cl	
6.23	-CH₃	-CH ₃	$-N$ CH_3	6-Cl	
6.24	-CH ₃	-CH ₃	-N_O	6-Cl	
6.25	-CH ₃	$-C_2H_5$	1-Naphthyl	6-C1	
6.26	-CH ₃	$-C_2H_5$	2-Naphthyl	6-Cl	
6.27	-CH ₃	$-C_2H_5$	$-N(CH_3)-c-C_6H_{11}$	6-Cl	
6.28	-CH ₃	$-C_2H_5$	-N(CH ₂ CH ₂ OCH ₃) ₂	6-Cl	
6.29	-CH ₃	$-C_2H_5$	N(CH ₂ CH ₃)C ₆ H ₅	6-Cl	
6.30	-CH ₃	-C ₂ H ₅	-N	6-Cl	

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Comp No.	o. R ₂	R ₃	В	R ₄	Phys. data
6.31	-CH ₃	-C ₂ H ₅	$-N$ CH_3	6-Cl	
6.32	-CH ₃	-C ₂ H ₅	-N_O	6-Cl	
6.33	-C ₂ H ₅	-C ₂ H ₅	1-Naphthyl	6-C1	181-183°
6.34	$-C_2H_5$	$-C_2H_5$	2-Naphthyl	6-C1	
6.35			$-N(CH_3)-c-C_6H_{11}$	6-C1	
6.36		$-C_2H_5$	-N(CH ₂ CH ₂ OCH ₃) ₂	6-C1	
6.37	$-C_2H_5$	$-C_2H_5$	N(CH ₂ CH ₃)C ₆ H ₅	6-C1	
6.38	-C ₂ H ₅	-C ₂ H ₅	-N	6-CI	
6.39	-C ₂ H ₅	-C ₂ H ₅	$-N$ CH_3	6-C1	
6.40	-C ₂ H ₅	-C ₂ H ₅	-N_O	6-Cl	
6.41	-сн ₂ -сн-с сн ₃ с	_	1-Naphthyl	6-Cl	
6.42	-сн ₂ -сн-с сн ₃ с		2-Naphthyl	6-Cl	
6.43	-сн ₂ -сн-с Сн ₃ с		-N(CH ₃)-c-C ₆ H ₁₁	6-Cl	
6.44	-сн ₂ -сн-с сн ₃ с		-N(CH ₂ CH ₂ OCH ₃) ₂	6-Cl	
6.45	-сн ₂ -сн-с сн ₃ с		N(CH ₂ CH ₃)C ₆ H ₅	6-Cl	

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Comp.	R ₂ R ₃	В	R ₄	Phys. data
6.46	-CH ₂ -CH-CH-CH ₂ - CH ₃ CH ₃		6-Cl	- Mand Specials and Market or Mills
6.47	-сн ₂ -сн-сн-сн ₂ - 	$-N$ CH_3 CH_3	6-Cl	
6.48	-сн ₂ -сн-сн-сн ₂ - I сн ₃ сн ₃	-N_O	6-Cl	·
6.49	-(CH ₂) ₄ -	1-Naphthyl	6-F	
6.50	$-(CH_2)_4$	2-Naphthyl	6-F	
6.51	-(CH ₂) ₄	$-N(CH_3)-c-C_6H_{11}$	6-F	
6.52	$-(CH_2)_4$	-N(CH ₂ CH ₂ OCH ₃) ₂	6-F	
6.53	-(CH ₂) ₄	$N(CH_2CH_3)C_6H_5$	6-F	
6.54	-(CH ₂) ₄	-N	6-F	
6.55	-(CH ₂) ₄	$-N$ CH_3	6-F	156-158°
6.56	-(CH ₂) ₄	-N_O	6-F	
6.57	-(CH ₂) ₃ -	1-Naphthyl	6-F	
6.58	-(CH ₂) ₃ -	2-Naphthyl	6-F	
6.59	-(CH ₂) ₃ -	$-N(CH_3)-c-C_6H_{11}$	6-F	
6.60	-(CH ₂) ₃ -	-N(CH ₂ CH ₂ OCH ₃) ₂	6-F	
6.61	-(CH ₂) ₃ -	$N(CH_2CH_3)C_6H_5$	6-F	

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Comp.	R ₂	R ₃	В	R ₄	Phys. data
6.62	-(CH ₂))3-	-N	6-F	
6.63	-(CH ₂))3-	$-N$ CH_3	6-F	
6.64	-(CH ₂))3-	-N	6-F	
6.65	-CH ₃	-CH ₃	1-Naphthyl	6-F	175-176°
6.66	-CH ₃	-CH ₃	2-Naphthyl	6-F	
6.67	$-CH_3$	-CH ₃	$-N(CH_3)-c-C_6H_{11}$	6-F	
6.68	-CH ₃	-CH ₃	-N(CH ₂ CH ₂ OCH ₃) ₂	6-F	
6.69	-CH ₃	-CH ₃	$N(CH_2CH_3)C_6H_5$	6-F	
6.70	-CH ₃	-CH ₃	-n	6-F	
6.71	-CH ₃	-CH ₃	$-N$ CH_3	6-F	156-158°
6.72	-CH ₃	-CH ₃	- N_O	6-F	
6.73	-CH ₃	$-C_2H_5$	1-Naphthyl	6-F	
6.74	-CH ₃		2-Naphthyl	6-F	
6.75	-CH ₃	$-C_2H_5$	$-N(CH_3)-c-C_6H_{11}$	6-F	
6.76	-CH ₃	$-C_2H_5$		6-F	
6.77	-CH ₃	$-C_2H_5$	N(CH ₂ CH ₃)C ₆ H ₅	6-F	

Comp.	R ₂	R ₃	В	R ₄	Phys. data
6.78	-CH ₃	-C ₂ H ₅	-N	6-F	and the second of the second o
6.79	-CH ₃	-C ₂ H ₅	$-$ N $ CH_3$	6-F	
6.80	-CH ₃	-C ₂ H ₅	-N_O	6-F	
6.81		$-C_2H_5$		6-F	
6.82			2-Naphthyl	6-F	
6.83			$-N(CH_3)-c-C_6H_{11}$	6-F	
6.84			-N(CH ₂ CH ₂ OCH ₃) ₂	6-F	
6.85	$-C_2H_5$	$-C_2H_5$	N(CH ₂ CH ₃)C ₆ H ₅	6-F	
6.86	-C ₂ H ₅	-C ₂ H ₅	-n	6-F	
6.87	-C ₂ H ₅	-C ₂ H ₅	—N ← CH ₃	6-F	
6.88	-C ₂ H ₅	-C ₂ H ₅	-N_O	6-F	
6.89	-СН ₂ -СН-4 СН ₃	_	1-Naphthyl	6-F	
6.90	-сн ₂ -сн-с сн ₃ 0	_	2-Naphthyl	6-F	
6.91	-сн ₂ -сн-с сн ₃ (-N(CH ₃)-c-C ₆ H ₁₁	6-F	

Phys. data

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Comp.	R ₂	R ₃	В	R ₄
6.92	1	н-сн-сн ₂ - ₁₃ сн ₃	-N(CH ₂ CH ₂ OCH ₃) ₂	6-F
6.93	- 1	н-сн-сн ₂ - ₁₃ сн ₃	N(CH ₂ CH ₃)C ₆ H ₅	6-F
6.94	1	н-сн-сн ₂ - -	-N	6-F
6.95		н-сн-сн ₂ - ₃ сн ₃	$-N$ CH_3	6-F
6.96		сн-сн ₂ - ₃ сн ₃	- N_O	6-F

Table 7

Comp. No.	R_2 R_3	B	R ₄	n	Phys. data
7.1	-(CH ₂) ₄ -	1-Naphthyl	2-CH ₃ ,6-CH ₃	2	
7.2	-(CH ₂) ₄ -	2-Naphthyl	2-CH ₃ ,6-CH ₃	2	
7.3	-(CH ₂) ₄ -	$-N(CH_3)-c-C_6H_{11}$	2-CH ₃ ,6-CH ₃	2	
7.4	-(CH ₂) ₄ -		2-CH ₃ ,6-CH ₃	2	
7.5	-(CH ₂) ₄ -	-N(CH2CH3)C6H5	2-CH ₃ ,6-CH ₃	2	
7.6	-(CH ₂) ₄ -	-N	2-CH ₃ ,6-CH ₃	2	
7.7	-(CH ₂) ₄ -	$-N$ CH_3	2-СН ₃ ,6-СН ₃	2	
7.8	-(CH ₂) ₄ -	-N_O	2-CH ₃ ,6-CH ₃	2	
7.9	-(CH ₂) ₄ -	1-Naphthyl	2-C ₂ H ₅ ,4-C ₂ H ₅ , 6-C ₂ H ₅	3	
7.10	-(CH ₂) ₄ -	2-Naphthyl	2-C ₂ H ₅ ,4-C ₂ H ₅ , 6-C ₂ H ₅	3	
7.11	-(CH ₂) ₄ -	$-N(CH_3)-c-C_6H_{11}$	2-C ₂ H ₅ ,4-C ₂ H ₅ , 6-C ₂ H ₅	3	
7.12	-(CH ₂) ₄ -	-N(CH ₂ CH ₂ OCH ₃) ₂	2-C ₂ H ₅ ,4-C ₂ H ₅ , 6-C ₂ H ₅	3	
7.13	-(CH ₂) ₄ -	-N(CH ₂ CH ₃)C ₆ H ₅	2-C ₂ H ₅ ,4-C ₂ H ₅ , 6-C ₂ H ₅	3	

Phys. data

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Comp No.	p. R ₂ R ₃	В	R ₄	n
7.14	-(CH ₂) ₄ -	-N	2-C ₂ H ₅ ,4-C ₂ H ₅ ,	3
			6-C ₂ H ₅	
7.15	-(CH ₂) ₄ -	$-N$ CH_3	2-C ₂ H ₅ ,4-C ₂ H ₅ ,	3
		J. 13	6-C ₂ H ₅	
7.16	-(CH ₂) ₄ -	-N_O	2-C ₂ H ₅ ,4-C ₂ H ₅ ,	3
_			6-C ₂ H ₅	
7.17	CH_3 CH_3	• •	2-CH ₃ ,6-CH ₃	2
7.18	CH ₃ CH ₃	• •	2-CH ₃ ,6-CH ₃	2
7.19	CH ₃ CH ₃	. 5. 0 11	2-CH ₃ ,6-CH ₃	2
7.20	CH_3 CH_3	. 2 2 3/4	2-CH ₃ ,6-CH ₃	2
7.21	CH ₃ CH ₃	$-N(CH_2CH_3)C_6H_5$	2-CH ₃ ,6-CH ₃	2
7.22	CH ₃ CH ₃	-n	2-CH ₃ ,6-CH ₃	2
7.23	CH ₃ CH ₃	—N ← CH ₃	2-CH ₃ ,6-CH ₃	2
7.24	CH ₃ CH ₃	-N	2-CH ₃ ,6-CH ₃	2
7.25	CH ₃ CH ₃	1-Naphthyl	2-C ₂ H ₅ ,4-C ₂ H ₅ , 6-C ₂ H ₅	3
7.26	CH ₃ CH ₃	2-Naphthyl	2-C ₂ H ₅ ,4-C ₂ H ₅ , 6-C ₂ H ₅	3
7.27	CH ₃ CH ₃	-N(CH ₃)-c-C ₆ H ₁₁	$2-C_2H_5, 4-C_2H_5,$	3

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	Comp. No.	R ₂	R ₃	В	R ₄	n	Phys. data
•					6-C ₂ H ₅		
,	7.28	CH ₃	CH ₃	-N(CH ₂ CH ₂ OCH ₃) ₂		3	
o i jegoro	7.29	CH ₃	CH ₃	-N(CH ₂ CH ₃)C ₆ H ₅ ,	-	.3	
,	7.30	СН3	CH ₃		2-C ₂ H ₅ ,4-C ₂ H ₅ ,	3	
					6-C ₂ H ₅		
	7.31	СН3	CH ₃	• 1	2-C ₂ H ₅ ,4-C ₂ H ₅ ,	3	
				ĊH ₃	6-C ₂ H ₅		
	7.32	CH ₃	CH ₃	-N_O	2-C ₂ H ₅ ,4-C ₂ H ₅ ,	3	
					6-C ₂ H ₅	•	
	7.33	-(CH	I ₂) ₃ -	1-Naphthyl	2-CH ₃ ,6-CH ₃	2	
	7.34	-(CH	I ₂) ₃ -	2-Naphthyl	2-CH ₃ ,6-CH ₃	2	
	7.35	-(CH	I ₂) ₃ -	$-N(CH_3)-c-C_6H_{11}$	2-CH ₃ ,6-CH ₃	2	
	7.36	-(CH	I ₂) ₃ -	-N(CH ₂ CH ₂ OCH ₃) ₂	2-CH ₃ ,6-CH ₃	2	
	7.37	-(CH	I ₂) ₃ -	-N(CH2CH3)C6H5	2-CH ₃ ,6-CH ₃	2	
	7.38	-(CF	I ₂) ₃ -	-N	2-CH ₃ ,6-CH ₃	2	
	7.39	-(CH	l ₂) ₃ -	-N CH ₃	2-CH ₃ ,6-CH ₃	2	
	7.40	-(CF	ł ₂) ₃ -	-N_O	2-CH ₃ ,6-CH ₃	2	

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Comp.	R ₂	R ₃	В	R ₄	n	Phys. data
7.41	-(CF	ł ₂) ₃ -	1-Naphthyl	2-C ₂ H ₅ ,4-C ₂ H ₅ , 6-C ₂ H ₅	3	
7.42	-(CF	I ₂) ₃ -	2-Naphthyl	2-C ₂ H ₅ ,4-C ₂ H ₅ , 6-C ₂ H ₅	3	
7.43	-(CF	I ₂) ₃ -	-N(CH ₃)-c-C ₆ H ₁₁	$2-C_2H_5, 4-C_2H_5,$ $6-C_2H_5$	3	
7.44	-(CH	I ₂) ₃ -	-N(CH ₂ CH ₂ OCH ₃) ₂		3	•
7.45	-(CH	l ₂) ₃ -	-N(CH ₂ CH ₃)C ₆ H ₅	2-C ₂ H ₅ ,4-C ₂ H ₅ , 6-C ₂ H ₅	3	
7.46	-(CH	2)3-	-N	2-C ₂ H ₅ ,4-C ₂ H ₅ ,	3	
			^	6-C ₂ H ₅		
7.47	-(CH	2)3-	$-N$ CH_3 2	2-C ₂ H ₅ ,4-C ₂ H ₅ ,	3	
			g	6-C ₂ H ₅		
7.48	-(CH ₂	2)3-	-N 0	2-C ₂ H ₅ ,4-C ₂ H ₅ ,	3	
,				6-C ₂ H ₅		

Table 8

$$(X)$$

$$O-CH_2-C-O-R_{15}$$

Comp. No.	X_2	R ₁₅
8.1	Cl	-CH(CH ₃)-C ₅ H ₁₁ -n
8.2	Cl	-CH(CH ₃)-CH ₂ OCH ₂ CH=CH ₂
8.3	Cl	Н
8.4	Cl	C_4H_9 -n

Table 9

$$\begin{array}{c|c}
E & COOR_{19} \\
 & N & N \\
 & R_{16} & R_{17}
\end{array}$$
(XI)

Comp. No.	R ₁₉	R ₁₆	R ₁₇	R ₁₈	E
9.1	CH ₃	Phenyl	2-Cl	Н	СН
9.2	CH ₃	Phenyl	2-Cl	4-Cl	СН
9.3	CH ₃	Phenyl	2-F	H	CH
9.4	CH ₃	2-Chlorophenyl	2-F	Н	CH
9.5	C_2H_5	CCl ₃	2-Cl	4-Cl	N
9.6	CH ₃	Phenyl	2-Cl	4-CF ₃	N
9.7	CH ₃	Phenyl	2-Cl	4-CF ₃	N



Formulation examples F1 to F10 for insecticidal and acaricidal active substances of the formula I (% = percent by weight)

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Example F1: Emulsion concentrates	a)	b)	c)
Active substance according to Tables 1-7	25%	•	50%
Calcium dodecylbenzenesulfonate	5%	8%	6%
Castor oil polyethylene glycol ether (36 mol EO)	5%	•	•
Tributylphenol polyethylene glycol ether			
(30 mol EO)	-	12%	4%
Cyclohexanone	-	15%	
Xylene mixture	65%	25%	

Mixing of finely ground active substance and additives gives an emulsion concentrate which on dilution with water gives emulsions of desired concentration.

Example F2: Solutions	a)	b)	c)	d)
Active substance according to			•	•
Tables 1-7	80%	10%	5%	95%
Ethylene glycol monomethyl				,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,
ether	20%	_	-	_
Polyethylene glycol (MW 400)	-	70%	-	-
N-Methylpyrrolid-2-one	-	20%	-	•
Epoxidized coconut oil	-	-	1%	5%
Petroleum spirit				- 10
(boiling range: 160-190°)	-	-	94%	_

Mixing of finely ground active substance and additives gives a solution suitable for use in the form of very small drops.

Example F3: Granules	a)	b)	c)	d)
Active substance according			ŕ	•
to Tab.1-7	5%	10%	8%	21%
Kaolin	94%	-	79%	54%
Highly disperse				
silica	1%	-	13%	7%
Attapulgite	-	90%	-	18%

The active substance is dissolved in dichloromethane, the solution is applied to the carrier mixture by spraying and the solvent is evaporated off in vacuo.

Example F4: Dust	a)	b)	
Active substance			
according to Tab. 1-7	2%	5%.	ės:
Highly disperse silica	1%	5%	
Talc	97%	-	
Kaolin	_	90%	

Mixing of active substance and carriers gives ready-to-use dusts.

Example F5: Wettable powder	a)	b)	c)
Active substance according to Tables 1-7	25%	50%	75%
Sodium ligninsulfonate	5%	5%	-
Sodium lauryl sulfate	3%	-	5%
Sodium diisobutylnaphthalenesulfonate	-	6%	10%
Octylphenol polyethylene glycol			
ether (7-8 mol EO)	-	2%	-
Highly disperse silica	5%	10%	10%
Kaolin	62%	27%	_

Active substance and additives are mixed and the mixture is ground in a suitable mill. Wettable powders are obtained which can be diluted with water to give suspensions of desired concentration.

Example F6: Emulsion concentrate

Active substance according to Tables 1-7	10%
Octylphenol polyethylene glycol	
ether (4-5 mol EO)	3%
Calcium dodecylbenzenesulfonate	3%
Castor oil polyethylene glycol	
ether (36 mol EO)	4%
Cyclohexanone	30%
Xylene mixture	50%

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Mixing of finely ground active substance and additives gives an emulsion concentrate which on dilution with water gives emulsions of desired concentration.

Example F7: Dust a) b)
Active substance
according to Tab. 1-7 8%
Talc 95% -

Kaolin - 92%

Application-ready dusts are obtained by mixing active substance and carrier and grinding the mixture in a suitable mill.

Example F8: Extruded granules

Active substance

according to Tab.1-7 10%
Sodium ligninsulfonate 2%
Carboxymethylcellulose 1%
Kaolin 87%

Active substance and additives are mixed, the mixture is ground, wetted with water, extruded and granulated, and the granules are dried in a stream of air.

Example F9: Coated granules

Active substance

according to Tab. 1-7 3%

Polyethylene glycol

(MW 200) 3% Kaolin 94%

Uniform application of the finely ground active substance to the kaolin, wetted with polyethylene glycol, in a mixer gives dust-free coated granules.

Example F10: Suspension concentrate

Active substance according to Tables 1-7

Ethylene glycol

10%

Nonylphenol polyethylene glycol ether	
(15 mol EO)	6%
Sodium ligninsulfonate	10%
Carboxymethylcellulose	1%
Aqueous formaldehyde solution (37%)	0.2%
Aqueous silicone oil emulsion (75%)	0.8%
Water	32%

Mixing of finely ground active substance and additives gives a suspension concentrate which on dilution with water gives suspensions of desired concentration.

Formulation Examples F11 to F18 for herbicidal active substances of the formula I (% = percent by weight)

F11. Emulsion concentrates	a)	b)	c)	d)
Active substance				
according to Tables 1-7	5 %	10 %	25 %	50 %
Calcium dodecylbenzenesulfonate	6 %	8 %	6 %	8 %
Castor oil polyglycol ether	4 %	-	4 %	4 %
(36 mol EO)				
Octylphenol polyglycol ether	-	4 %	-	2 %
(7-8 mol EO)				•
Cyclohexanone	-	-	10 %	20 %
Aromatic hydrocarbon mixture	85 %	78 %	55 %	16 %
C ₉ -C ₁₂				

From such concentrates, by dilution with water, it is possible to prepare emulsions of any desired concentration.

F12. Solutions	a)	b)	c)	d)
Active substance				
according to Tables 1-7	5 %	10 %	50 %	90 %
Dipropylene glycolmethyl ether	-	20 %	20 %	•
Polyethylene glycol MW 400	20 %	10 %	-	-
N-Methyl-2-pyrrolidone	-	-	30 %	10 %
Aromatic hydrocarbon mixture	75 %	60 %	- ,	-

C₉-C₁₂

The solutions are suitable for use in the form of very small drops.

F13. Wettable powders	a)	b)	c)	d)
Active substance		·	•	_,
according to Tables 1-7	5 %	25 %	50 %	80 %
Sodium ligninsulfonate	4 %	-	3 %	•
Sodium lauryl sulfate	2 %	3 %	-	4 %
Sodium diisobutylnaphthalene-	-	6 %	5 %	6%
sulfonate				
Octylphenol polyglycol ether	-	1 %	2 %	_
(7-8 mol EO)				
Highly disperse silica	1 %	3 %	5 %	10 %
Kaolin	88 %	62 %	35 %	-

The active substance is mixed thoroughly with the additives and the mixture is ground thoroughly in a suitable mill. Wettable powders are obtained which can be diluted with water to give suspensions of any desired concentration.

F14. Coated granules	a)	b)	c)
Active substance	-,	• ,	Ο,
according to Tables 1-7	0.1 %	5 %	15 %
Highly disperse silica	0.9 %	2 %	2 %
Inorganic carrier material	99.0 %	93 %	83 %
(Ø 0.1 - 1 mm)			
such as, for example, CaCO ₃ or SiO ₂			

The active substance is dissolved in methylene chloride, the solution is applied to the carrier by spraying, and the solvent is subsequently evaporated off in vacuo.

F15. Coated granules	a)	b)	c)
Active substance	·	,	-,
according to Tables 1-7	0.1 %	5 %	15 %
Polyethylene glycol MW 200	1.0 %	2 %	3 %
Highly disperse silica	0.9 %	1 %	2 %

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Inorganic carrier material 98.0 % 92 % 80 % (Ø 0.1 - 1 mm) such as, for example, CaCO₃ or SiO₂

The finely ground active substance is applied uniformly, in a mixer, to the carrier material wetted with polyethylene glycol. In this way, dust-free coated granules are obtained.

F. 16's 12's 2'No fee about make an a color			
a)	b)	c)	d)
0.1 %	3 %	5 %	15 %
1.5 %	2 %	3 %	4 %
1.4 %	2 %	2 %	2 %
97.0 %	93 %	90 %	79 %
	a) 0.1 % 1.5 % 1.4 %	a) b) 0.1 % 3 % 1.5 % 2 % 1.4 % 2 %	a) b) c) 0.1 % 3 % 5 % 1.5 % 2 % 3 % 1.4 % 2 % 2 %

The active substance is mixed with the additives, and the mixture is ground and wetted with water. This mixture is extruded and subsequently dried in a stream of air.

F17. Dusts	a)	b)	c)
Active substance			
according to Tables 1-7	0.1 %	1 %	5 %
Talc	39.9 %	49 %	35 %
Kaolin	60.0 %	50 %	60 %

Application-ready dusts are obtained by mixing the active substance with the carriers and grinding the mixture on a suitable mill.

F18. Suspension concentrates	a)	b)	c)	d)
Active substance				
according to Tables 1-7	3 %	10 %	25 %	50 %
Ethylene glycol	5 %	5 %	5 %	5 %
Nonylphenol polyglycol ether	-	1 %	2 %	-
(15 mol EO)				
Sodium ligninsulfonate	3 %	3 %	4 %	5 %
Carboxymethylcellulose	1 %	1 %	1 %	1 %
37% aqueous formaldehyde solution	0.2 %	0.2 %	0.2 %	0.2 %
Silicone oil emulsion	0.8 %	0.8 %	0.8 %	0.8 %

Water

87 %

79 %

62 %

38 %

The finely ground active substance is intimately mixed with the additives. In this way a suspension concentrate is obtained which can be diluted with water to give suspensions of any desired concentration.

Formulation Examples F19 to F26 for mixtures of herbicides of the formula I and safeners of the formula X or XI (% = percent by weight)

F19. Emulsion concentrates	a)	b)	c)	d)
Active substance mixture	5 %	10 %	25 %	50 %
Calcium dodecylbenzenesulfonate	6 %	8 %	6 %	8 %
Castor oil polyglycol ether	4 %	-	4 %	4 %
(36 mol EO)			. , ,	. ,0
Octylphenol polyglycol ether	•	4 %	-	2 %
(7-8 mol EO)				2 70
Cyclohexanone	-	•	10 %	20 %
Aromatic hydrocarbon mixture	85 %	78 %	55 %	16 %
C ₉ -C ₁₂			22 70	10 70

From such concentrates it is possible by dilution with water to prepare emulsions of any desired concentration.

F20. Solutions	a)	b)	c)	d)
Active substance mixture	5 %	10 %	50 %	90 %
Dipropylene glycol methyl ether	-	20 %	20 %	-
Polyethylene glycol MW 400	20 %	10 %	-	_
N-Methyl-2-pyrrolidone	-	•	30 <i>%</i>	10 %
Aromatic hydrocarbon mixture	75 %	60 %	-	-
C ₉ -C ₁₂				

The solutions are suitable for use in the form of very small drops.

F21. Wettable powders	a)	b)	c)	d)
Active substance mixture	5 %	25 %	50 %	80 %
Sodium ligninsulfonate	4 %	-	3 %	-

Sodium lauryl sulfate	2 9	% 3%	-	4 %
Sodium diisobutylnaphthalene-				
sulfonate	-	6 %	5 %	6 %
Octylphenol polyglycol ether	•	1 %	2 %	-
(7-8 mol EO)				
Highly disperse silica	1 9	% 3 %	5 %	10 %
Kaolin	88 %	62 %	.35.%,	appearance of

The active substance is mixed thoroughly with the additives and the mixture is ground thoroughly in a suitable mill. Wettable powders are obtained which can be diluted with water to give suspensions of any desired concentration.

F22. Coated granules	a)	b)	c)
Active substance mixture	0.1 %	5 %	15 %
Highly disperse silica	0.9 %	2 %	2 %
Inorganic carrier material	99.0 %	93 %	83 %
(Ø 0.1 - 1 mm)			
such as, for example, CaCO ₃ or SiO ₂			

The active substance is dissolved in methylene chloride, the solution is applied to the carrier by spraying and the solvent is subsequently evaporated off in vacuo.

F23. Coated granules	a)	b)	c)
Active substance mixture	0.1 %	5 %	15 %
Polyethylene glycol MW 200	1.0 %	2 %	3 %
Highly disperse silica	0.9 %	1 %	2 %
Inorganic carrier material	98.0 %	92 %	80 %
(Ø 0.1 - 1 mm)			
such as, for example, CaCO ₃ or SiO ₂			

The finely ground active substance is applied uniformly, in a mixer, to the carrier material wetted with polyethylene glycol. In this way, dust-free coated granules are obtained.

F24. Extruded granules	a)	b)	c)	d)
Active substance mixture	0.1 %	3 %	5 %	15 %
Sodium ligninsulfonate	1.5 %	2 %	3 %	4 %

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Carboxymethylcellulose	1.4 %	2 %	2 %	2 %
Kaolin	97.0 %	93 %	90 %	79 <i>%</i>

The active substance is mixed with the additives, ground and wetted with water. This mixture is extruded and then dried in a stream of air.

F25. Dusts	a)	b)	c)
Active substance mixture	0.1 %	1 %	5 %
Talc	39.9 %	49 %	35 %
Kaolin	60.0 %	50 <i>%</i>	60 %

Application-ready dusts are obtained by mixing the active substance with the carriers and grinding the mixture in a suitable mill.

F26. Suspension concentrates	a)	b)	c)	d)
Active substance mixture	3 %	10 %	25 %	50 %
Ethylene glycol	5 %	5 %	5 %	5 %
Nonylphenol polyglycol ether	-	1 %	2 %	-
(15 mol EO)				
Sodium ligninsulfonate	3 %	3 %	4 %	5 %
Carboxymethylcellulose	1 %	1 %	1 %	1 %
37% aqueous formaldehyde	0.2 %	0.2 %	0.2 %	0.2 %
solution				
Silicone oil emulsion	0.8 %	0.8 %	0.8 %	0.8 %
Water	87 %	79 %	62 %	38 %

The finely ground active substance is intimately mixed with the additives. In this way, a suspension concentrate is obtained which can be diluted with water to give suspensions of any desired concentration.

<u>Biological examples</u> (% = per cent by weight, unless otherwise indicated)

A. Insecticidal action

Example B1: Action against Myzus persicae

Pea seedlings are infected with Myzus persicae, subsequently sprayed with a spray

mixture comprising 400 ppm of active ingredient and then incubated at 20°. The test is evaluated after 3 and 6 days. The percentage reduction in the population (% action) is determined by comparing the number of dead aphids on the treated plants with those on untreated plants.

Compounds of Tables 1 to 7 exhibit a good action in this test.

Example B2: Action against Nilaparvata lugens

Rice plants are treated with an aqueous emulsion spray liquor comprising 400 ppm of active ingredient and, after the spray coating has dried on, are populated with stage 2 and 3 cicada larvae. 21 days later, the percentage reduction in the population (% action) is determined by comparing the number of surviving cicadas on the treated plants with those on untreated plants.

Compounds of Tables 1 to 7 exhibit a good action in this test.

Example B3: Action against Nilaparvata lugens (systemic)

Pots containing rice plants are placed in an aqueous emulsion solution comprising 400 ppm of active ingredient. The plants are then populated with stage 2 and 3 larvae. 6 days later, the percentage reduction in the population (% action) is determined by comparing the number of cicadas on the treated plants with those on untreated plants.

Compounds of Tables 1 to 7 exhibit a good action in this test.

B. Acaricidal action

Example B4: Action against Boophilus microplus

Fully satiated adult female ticks are stuck onto a PVC plate and covered with a cottonwool pad, and 10 ml of aqueous test solution comprising 125 ppm of active ingredient are poured over the animals. The cottonwool pad is removed, and the ticks are incubated for 4 weeks for oviposition. The action is evident either in the female, as mortality or sterility, or in the eggs, as ovicidal action.

Compounds of Tables 1 to 7 exhibit a good action in this test.

Example B5: Action against Tetranychus urticae

Young bean plants are populated with a mixed population of Tetranychus urticae, sprayed 1 day later with an aqueous emulsion spray liquor comprising 400 ppm of active ingredient, incubated at 25° for 6 days and subsequently evaluated. The percentage reduction in the population (% action) is determined by comparing the number of dead

eggs, larvae and adults on the treated plants with those on untreated plants. Compounds of Tables 1 to 7 exhibit a good action in this test. An action of more than 80% is shown, in particular, by the compounds nos. 1.1 to 1.9, 1.11 to 1.28, 1.33 to 1.40, 1.44 to 1.49, 1.51, 1.53, 1.55 to 1.61, 2.2, 2.6 to 2.14, 2.16, 2.17, 2.20, 2.21, 3.1, 5.15, 5.47, 5.80, 5.88, 5.107, 5.110, 5.112, 5.122, 6.2 to 6.4 and 6.7.

C. Herbicidal action

Example B6: Herbicidal action before emergence of the plants (preemergence)

Monocotyledon and dicotyledon test plants are sown in standard soil in plastic pots. Directly after sowing, the test substances are sprayed on in an aqueous suspension prepared from a 25 % wettable powder (Example F13, b)), at a rate of 2 kg of AS/ha (500 l of water/ha). The test plants are subsequently grown in the greenhouse under optimum conditions. After a test duration of 3 weeks, the experiment is evaluated using a 9-point rating scale (1 = complete damage, 9 = no action). Ratings from 1 to 4 (especially 1 to 3) denote a good to very good herbicidal action.

In this experiment, compounds of Tables 1-7 exhibit a strong herbicidal action, as can be seen, for example, from the results presented in Table 10.

Table 10

Compound No.	Avena	Setaria	Sinapis	Stellaria	
1.5	3	1	3	8	
1.9	2	2	2	7	
1.13	2	1	3	9	
1.14	2	1	2	9	
1.19	2	1	2	5	
1.21	2	1	3	7	
1.24	3	1	3	7	
1.25	3	1	3	7	
1.26	1	1	2	8	
1.27	3	1	3	7	
1.33	2	1	2	9	
1.35	2	1	1	6	
4.15	1	1	9	9	

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4.16	1	1	9	8
4.22	1	2	9	9

Example B7: Post-emergence herbicidal action (contact herbicide)

Monocotyledon and dicotyledon test plants are grown in standard soil in the greenhouse in plastic pots and, at the 4- to 6-leaf stage, are sprayed with an aqueous suspension of the test substances of the formula I, prepared from a 25 % wettable powder (Example F13, b)), at a rate of 2000 g of AS/ha (500 l of water/ha). The test plants are subsequently grown on in the greenhouse under optimum conditions. After a test period of about 18 days, the experiment is evaluated using a 9-point rating scale (1 = complete damage, 9 = no action). Ratings from 1 to 4 (especially 1 to 3) denote a good to very good herbicidal action.

In this experiment, compounds of Tables 1-7 exhibit a strong herbicidal action, as can be seen, for example, from the results presented in Table 11.

Table 11

Compound No.	Avena	Setaria	Sinapis	Stellaria	
1.5	1	2	2	5	
1.9	1	1	2	4	
1.13	1	2	2	3	
1.14	1	2	3	5	
1.19	1	2	2	4	
1.21	2	3	3	6	
1.24	1	1	3	4	
1.25	1	1	3	4	
1.26	1	1	2	3	
1.27	1	1	2	7	
1.33	1	1	2	4	
1.35	1	1	2	5	
4.15	1	3	3	9	
4.16	1	2	3	7	
4.22	1	2	2	9	

D. Safener effect

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Example B8: Safener effect

Under greenhouse conditions, the test plants are grown in plastic pots to the 4-leaf stage. At this stage, both the herbicide alone and the mixtures of the herbicide with the test substances to be tested as safeners are applied to the test plants. Application takes place in the form of an aqueous suspension of the test substances, prepared from a 25 % wettable powder (Example F21, b)), with 500 l of water/ha. 3 weeks after application, the phytotoxic action of the herbicide on the crop plants, for example maize and cereal, is evaluated using a percentage scale. 100 % denotes that the test plant has died, while 0 % denotes no phytotoxic effect.

This experiment shows that, with compounds of the formula X or XI, for example with the compound no. 8.1, the damage caused to crop plants, for example maize or cereal plants, by herbicide compounds of the formula I, for example by the compound no. 1.13, can be substantially reduced.

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WHAT IS CLAIMED IS:

1. A compound of the formula

in which

$$R_1$$
 is the group $(R_4)_n$, $N = (R_5)_m$ or $(R_4)_n$;

the substituents R₄ independently of one another are halogen, nitro, cyano, C₁-C₄alkyl, C_1 - C_4 haloalkyl, C_1 - C_{10} alkoxy, C_1 - C_4 haloalkoxy, C_3 - C_6 alkenyloxy,

C₁-C₄alkoxy-C₂-C₄alkoxy, C₃-C₆alkynyloxy, C₁-C₄alkylcarbonyl, C₁-C₄alkoxycarbonyl, C_1 - C_4 alkylthio, C_1 - C_4 alkylsulfinyl, C_1 - C_4 alkylsulfonyl, amino, C_1 - C_4 alkylamino or $di(C_1-C_4alkyl)amino;$

$$R_5$$
 is the group $-x_1$ or $-x_1$ (R_6) q

n is 0, 1, 2, 3 or 4;

m is 0 or 1, and the sum of m and n is 0, 1, 2, 3 or 4;

q is 0, 1, 2 or 3;

 X_1 is oxygen, sulfur, -CH₂- or -N(R₇)-;

the substituents R_6 independently of one another are C_1 - C_4 alkyl, halogen, C_1 - C_4 haloalkyl, C_1 - C_4 alkoxy, C_1 - C_4 haloalkoxy, nitro, cyano, C_1 - C_4 alkoxycarbonyl, amino,

 C_1 - C_4 alkylamino or di(C_1 - C_4 alkyl)amino;

R₇ is hydrogen, C₁-C₄alkyl, formyl or C₁-C₄alkylcarbonyl;

R₂ and R₃ independently of one another are hydrogen, C₁-C₈alkyl, C₃-C₈alkenyl,

 C_3 - C_8 alkynyl, C_1 - C_8 alkoxyalkyl, C_1 - C_8 alkylthioalkyl, C_3 - C_8 cycloalkyl or substituted or unsubstituted aryl; or

R₂ and R₃ together form the bivalent radical of a saturated or unsaturated, unsubstituted or substituted, mono-, bi-, tri- or polycyclic system, which system can optionally contain, in positions not adjacent to the nitrogen atoms shown in formula I, one or more ring hetero atoms;

G is the group -CO-A (a) or -SO₂-B (b);

A is C₁-C₈alkyl substituted by nitro, cyano, Si(C₁-C₄alkyl)₃, amino, C₁-C₄alkylamino,

di(C_1 - C_4 alkyl)amino, substituted or unsubstituted benzyloxy or a group $-CH-NR_{08}R_{09}$, $COOR_{010}$

or A is C_3 - C_8 cycloalkyl which can if desired contain as heteroatom oxygen, sulfur or nitrogen and is substituted by C_1 - C_4 alkyl, C_1 - C_4 alkoxy, C_1 - C_4 alkylsulfinyl, C_1 - C_4 alkylsulfonyl or substituted or unsubstituted phenyl, or A is C_3 - C_8 cycloalkyloxy which can if desired contain as heteroatom oxygen, sulfur or nitrogen and is substituted by C_1 - C_4 alkyl, or A is adamantyl, naphthyl, naphthyl substituted by halogen, C_1 - C_4 alkyl, C_1 - C_4 haloalkyl or C_1 - C_4 alkoxy, or A is a group - $(CR_8R_9)_p$ - $(Y)_o$ -CO- $(Z)_r$ - R_{10} , - $(CR_8R_9)_p$ - R_{11} or - $N(R_{10})R_{13}$;

R₀₈, R₀₉ and R₀₁₀ independently of one another are hydrogen or C₁-C₆alkyl; R₈ and R₉ independently of one another are hydrogen, C₁-C₆alkyl or phenyl; R₁₀ is hydrogen, C₁-C₁₀alkyl, C₁-C₁₀alkyl substituted by halogen, C₁-C₄alkoxy, C₁-C₄haloalkoxy, C₁-C₄alkylthio, C₃-C₁₂cycloalkyl, cyano, aryl or aryloxy, C₃-C₈cycloalkyl, C₃-C₈cycloalkyl substituted by halogen or C₁-C₄alkyl, C₃-C₁₀alkenyl, C₃-C₁₀alkynyl, C₃-C₁₀haloalkenyl, C₃-C₁₀haloalkynyl, aryl, aryl substituted by halogen, C₁-C₄alkyl, C₁-C₄haloalkyl, C₁-C₆alkoxy, C₁-C₄haloalkoxy, C₁-C₄alkylthio, nitro, cyano, phenoxy or halo-substituted phenoxy:

o and r independently of one another are 0 or 1;

Y and Z independently of one another are oxygen or NR_{12} ; p is 1 to 10;

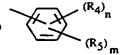
 R_{11} is C_3 - C_8 cycloalkyl, C_3 - C_8 cycloalkyl substituted by halogen or C_1 - C_4 alkyl, aryloxy, aryloxy substituted by halogen, C_1 - C_4 alkyl, C_1 - C_4 haloalkyl, C_1 - C_4 alkoxy, C_1 - C_4 haloalkoxy, C_1 - C_4 alkylthio, nitro, cyano, phenoxy or halo-substituted phenoxy, benzyloxy substituted by halogen, C_1 - C_4 alkyl, C_1 - C_4 haloalkyl, C_1 - C_4 alkoxy, C_1 - C_4 haloalkoxy, C_1 - C_4 alkylthio, nitro, cyano, phenoxy or halo-substituted phenoxy, norbornyl or adamantyl, where aryloxy with the exception of phenoxy can in each case be any desired aryloxy group;

 R_{12} is hydrogen, C_1 - C_4 alkyl, C_3 - C_8 cycloalkyl, C_3 - C_{10} alkenyl or C_3 - C_{10} alkynyl; R_{13} is C_3 - C_8 cycloalkyl, C_3 - C_8 cycloalkyl substituted by halogen or C_1 - C_4 alkyl, or cyano- C_1 - C_6 alkyl;

B is C_1 - C_{10} alkyl substituted by C_1 - C_4 alkoxy, C_1 - C_6 alkoxy- C_1 - C_4 alkoxy, C_1 - C_4 alkylthio, C_1 - C_4 haloalkylthio, C_1 - C_4 alkylsulfinyl, C_1 - C_4 alkylsulfonyl, C_1 - C_4 alkylsulfonyloxy, C_1 - C_4 alkylsulfonyloxy, C_1 - C_4 alkylcarbonyloxy, C_1 - C_4 alkylcarbonyloxy, C_1 - C_4 alkyl)amino, C_3 - C_8 cycloalkyl, aryl, aryloxy, arylthio, arylsulfonyl, arylsulfonyloxy, arylcarbonyl or pyridyl, it being possible for the aryl and pyridyl groups if desired to be substituted by halogen, C_1 - C_4 alkyl,

C₁-C₄haloalkyl, C₁-C₄alkoxy, C₁-C₄haloalkoxy, C₁-C₄alkylthio, nitro, cyano, phenoxy, halo-substituted phenoxy, phenylthio or halo-substituted phenylthio, or B is C₃-C₁₀alkenyl, C₃-C₁₀haloalkenyl, C₃-C₁₀alkynyl, C₃-C₁₀haloalkynyl, C₃-C₈cycloalkyl, C₃-C₈cycloalkyl substituted by halogen or C₁-C₄alkyl, benzyl, benzyl substituted by halogen, C₁-C₄alkyl, C₁-C₄haloalkyl or C₁-C₄alkoxy, naphthyl, naphthyl substituted by halogen or nitro, or B is a group -N(R₂₁)R₂₂: R₂₁ is C₁-C₁₀alkyl substituted by C₁-C₄alkoxy, C₁-C₆alkoxy-C₁-C₄alkoxy, C₁-C₄haloalkoxy, C₁-C₄alkylthio, C₁-C₄haloalkylthio, C₁-C₄alkylsulfinyl, C₁-C₄alkylsulfonyl, C₁-C₄alkylsulfonyloxy, C₁-C₄alkylcarbonyl, C₁-C₄alkoxycarbonyl, C₁-C₄alkylcarbonyloxy, di(C₁-C₄alkyl)amino, C₃-C₈cycloalkyl, aryl, aryloxy, arylthio, arylsulfonyl, arylsulfinyl, arylsulfonyloxy, arylcarbonyl or pyridyl, it being possible for the aryl and pyridyl groups if desired to be substituted by halogen, C₁-C₄alkyl, C₁-C₄haloalkyl, C₁-C₄alkoxy, C₁-C₄haloalkoxy, C₁-C₄alkylthio, nitro, cyano, phenoxy, halo-substituted phenoxy, phenylthio or halo-substituted phenylthio, or R₂₁ is C₃-C₂₀alkenyl, C₃-C₂₀haloalkenyl, C₃-C₂₀alkynyl, C₃-C₂₀haloalkynyl, C₃-C₈cycloalkyl, C₃-C₈cycloalkyl substituted by halogen or C₁-C₄alkyl, benzyl or aryl, it being possible for the benzyl and aryl groups if desired to be substituted by halogen, C₁-C₄alkyl, C₁-C₄haloalkyl, C₁-C₄alkoxy, C₁-C₄haloalkoxy, C₁-C₄alkylthio, nitro, cyano, benzoyl, halo-substituted benzoyl, phenoxy or halo-substituted phenoxy; R_{22} is as defined for R_{21} or else alternatively is C_1 - C_{10} alkyl; or R₂₁ and R₂₂, together with the nitrogen atom to which they are attached, form an unsubstituted or mono- to trisubstituted, nonaromatic, mono- or bicyclic heterocyclic ring in which the substituents can be C₁-C₄alkyl, C₁-C₄alkyl substituted by halogen, C₁-C₄alkoxy, phenyl, benzodioxoyl or trifluoromethylphenyl, C₁-C₄alkoxy, C₁-C₄alkoxycarbonyl, C₁-C₄alkylcarbonyl, phenyl, phenyl substituted by halogen, C₁-C₄alkyl, C₁-C₄alkoxy, nitro or trifluoromethyl, pyridyl, pyrimidinyl or formyl, and salts and diastereomers of the compounds of the formula I, with the proviso that, if R₂₁ and R₂₂ are alkyl, at least one of these radicals is substituted alkyl.

2. A compound according to claim 1, in which R₁ is the group



3. A compound according to claim 2, in which R_4 is fluorine, chlorine or C_1 - C_4 alkyl; n is 1, 2 or 3; and m is 0.

- 4. A compound according to claim 3, in which R_4 is fluorine, chlorine or methyl; and n is 2 or 3.
- 5. A compound according to claim 1, in which R_2 and R_3 are C_1 - C_6 alkyl; or R_2 and R_3 together form a substituted or unsubstituted alkylene chain -(CH₂)₅-, -(CH₂)₄- or -(CH₂)₃-.
- 6. A compound according to claim 1, in which G is the group -CO-A (a).
- 7. A compound according to claim 6, in which A is a group - $(CR_8R_9)_p$ - $(Y)_o$ -CO- $(Z)_r$ - R_{10} , - $(CR_8R_9)_p$ - R_{11} or - $N(R_{10})R_{13}$, or substituted or unsubstituted naphthyl.
- 8. A compound according to claim 7, in which R_8 and R_9 independently of one another are hydrogen or methyl; and p is 1 or 2.
- 9. A compound according to claim 8, in which R₈ and R₉ are methyl.
- 10. A compound according to claim 7, in which R_{10} is C_1 - C_8 alkyl, C_3 - C_6 cycloalkyl, phenyl, phenyl substituted by halogen, or is naphthyl; R_{11} is C_3 - C_8 cycloalkyl, benzyloxy, benzyloxy substituted by halogen, C_1 - C_4 alkyl, C_1 - C_4 haloalkyl or phenoxy, or is naphthyloxy; and R_{13} is C_3 - C_7 cycloalkyl or cyano- C_1 - C_6 alkyl.
- 11. A compound according to claim 7, in which Y and Z are NR₁₂.
- 12. A compound according to claim 11, in which R_{12} is hydrogen or C_1 - C_4 alkyl.
- 13. A compound according to claim 7, in which R_8 and R_9 independently of one another are hydrogen or methyl; p is 1 or 2; R_{10} is C_1 - C_4 alkyl, C_3 - C_6 cycloalkyl, phenyl, phenyl substituted by halogen, or is naphthyl; R_{11} is C_3 - C_6 cycloalkyl, benzyloxy, benzyloxy substituted by halogen, C_1 - C_4 alkyl, C_1 - C_4 haloalkyl or phenoxy, or is naphthyloxy; Y and Z are NR_{12} ; R_{12} is hydrogen or C_1 - C_4 alkyl; and R_{13} is C_3 - C_8 cycloalkyl or cyano- C_1 - C_6 alkyl.
- 14. A compound according to claim 1, in which R_1 is the group $(R_5)_m$; R_4 is

fluorine, chlorine or C_1 - C_4 alkyl; n is 1, 2 or 3; m is 0; R_2 and R_3 are C_1 - C_6 alkyl; or R_2

and R_3 together form a substituted or unsubstituted alkylene chain - $(CH_2)_5$ -, - $(CH_2)_4$ - or - $(CH_2)_3$ -; G is the group -(CO-A) (a); A is a group - $(CR_8R_9)_p$ - $(Y)_o$ - $(CO-(Z)_r$ - $(CR_8R_9)_p$ - $(Y)_o$ - $(CO-(Z)_r$ - $(CR_8R_9)_p$ - $(Y)_o$ - $(CR_8R_9)_p$ - $(Y)_o$ - $(CO-(Z)_r$ - $(CR_8R_9)_p$ - $(Y)_o$ - $(CO-(Z)_r$ -

- 15. A compound according to claim 1, in which R_1 is the group $(R_4)_n$; and R_5 is 0.
- 16. A compound according to claim 15, in which R₄ is C₁- or C₂alkyl; and n is 1, 2 or 3.
- 17. A compound according to claim 16, in which R_4 is C_1 or C_2 alkyl in positions 2, 4 and 6 relative to the phenyl linkage site; and n is 3.
- 18. A compound according to claim 1, in which R_1 is the group $(R_4)_n$; m is 0;

 R_4 is C_1 - or C_2 alkyl; n is 1, 2 or 3; R_2 and R_3 are C_1 - C_6 alkyl; or R_2 and R_3 together form a substituted or unsubstituted alkylene chain -(CH₂)₅-, -(CH₂)₄- or -(CH₂)₃-.

19. A compound according to claim 1, in which G is a group -CO-A (a); and A is C_1 - C_8 alkyl substituted by nitro, cyano, $Si(C_1$ - C_4 alkyl)₃, amino, C_1 - C_4 alkylamino, di(C_1 - C_4 alkyl)amino, substituted or unsubstituted benzyloxy or a group $-CH-NR_{08}R_{09}$, $COOR_{010}$

or A is C_3 - C_8 cycloalkyl, which can if desired contain as heteroatom oxygen, sulfur or nitrogen and is substituted by C_1 - C_4 alkyl, C_1 - C_4 alkoxy, C_1 - C_4 alkylsulfinyl, C_1 - C_4 alkylsulfonyl or substituted or unsubstituted phenyl, or A is C_3 - C_8 cycloalkyloxy which can if desired contain as heteroatom oxygen, sulfur or nitrogen and is unsubstituted or substituted by C_1 - C_4 alkyl, or A is a group - $(CR_8R_9)_p$ - $(Y)_o$ -CO- $(Z)_r$ - R_{10} or - $(CR_8R_9)_p$ - R_{11} .

٦,

20. A compound according to claim 1, in which R₁ is the group

$$(R_4)_n$$
 ; R_4 is

C₁- or C₂alkyl; n is 1, 2 or 3; m is 0; R₂ and R₃ are C₁-C₆alkyl; or R₂ and R₃ together form a substituted or unsubstituted alkylene chain -(CH₂)₅-, -(CH₂)₄- or -(CH₂)₃-; G is the group -CO-A (a); and A is C₁-C₈alkyl substituted by nitro, cyano, Si(C₁-C₄alkyl)₃, amino, C₁-C₄alkylamino, di(C₁-C₄alkyl)amino, substituted or unsubstituted benzyloxy or a group -CH-NR₀₈R₀₉, or A is C₃-C₈cycloalkyl which can if desired contain as heteroatom COOR₀₁₀

oxygen, sulfur or nitrogen and is substituted by C_1 - C_4 alkyl, C_1 - C_4 alkoxy, C_1 - C_4 alkylsulfinyl, C_1 - C_4 alkylsulfonyl or substituted or unsubstituted phenyl, or A is C_3 - C_8 cycloalkyloxy which can if desired contain as heteroatom oxygen, sulfur or nitrogen and is unsubstituted or substituted by C_1 - C_4 alkyl, or A is a group - $(CR_8R_9)_p$ - $(Y)_o$ -CO- $(Z)_r$ - R_{10} or - $(CR_8R_9)_p$ - R_{11} .

21. A process for the preparation of a compound of the formula

$$\begin{array}{c}
R_3 \\
N \\
R_2
\end{array}$$

$$\begin{array}{c}
N \\
R_1
\end{array}$$
(I),

in which R_1 , R_2 and R_3 are as defined in claim 1, G is the group -CO-A (a) and A is as defined in claim 1 with the exception of the group -N(R_{10}) R_{13} , which comprises reacting a compound of the formula

$$R_3$$
 N
 R_2
 N
 R_1
 R_1
 R_1
 R_1

in which R₁, R₂ and R₃ are as defined, with a compound of the formula

$$\begin{array}{c}
O \\
II \\
A - C - E_1
\end{array} (III)$$

in which A is as defined and E₁ is a leaving group.

22. A process for the preparation of a compound of the formula

$$\begin{array}{c|c}
R_3 & O - G \\
R_2 & R_1
\end{array}$$
(I),

in which R₁, R₂ and R₃ are as defined in claim 1, G is the group -CO-A (a), and A is -N(R_{10}) R_{13} , where R_{10} and R_{13} are as defined in claim 1 with the exception of R_{10} as hydrogen, which comprises reacting a compound of the formula

$$R_3$$
 R_2
 N
 R_1
 R_1
 R_1
 R_1

in which
$$R_1$$
, R_2 and R_3 are as defined, with a compound of the formula
$$E_1 - \overset{O}{C} - \overset{R_{10}}{N_{13}}$$
 (IV),

in which R_{10} and R_{13} are as defined and E_1 is a leaving group.

23. A process for the preparation of a compound of the formula

$$R_3$$
 R_2
 R_1
 R_1
 R_1
 R_1
 R_2

in which R₁, R₂ and R₃ are as defined in claim 1, G is the group -CO-A (a), and A is -NHR₁₃, where R₁₃ is as defined in claim 1, which comprises reacting a compound of the formula

$$R_3$$
 R_2
 R_1
 R_1
 R_1
 R_1
 R_1

in which R₁, R₂ and R₃ are as defined, with an isocyanate of the formula

$$R_{13}$$
-N=C=O (V),

in which R₁₃ is as defined.

24. A process for the preparation of a compound of the formula

$$\begin{array}{c|c}
R_3 & O - G \\
R_2 & R_1
\end{array}$$
(I),

in which R_1 , R_2 and R_3 are as defined in claim 1, G is the group -SO₂-B (b), and B is as defined in claim 1, which comprises reacting a compound of the formula

$$R_3$$
 N
 R_2
 N
 R_1
 R_1
 R_1
 R_1

in which R_1 , R_2 and R_3 are as defined in claim 1, with a compound of the formula $B-SO_2-E_1$ (VI),

in which B is as defined and E₁ is a leaving group, if desired in a solvent and in the presence or absence of a base.

- 25. A pesticidal composition comprising, in addition to customary formulation auxiliaries, at least one compound of the formula I according to claim 1 as active substance.
- 26. The use of a pesticidal composition according to claim 25 for controlling insects, representatives of the order Acarina or broad-leaved weeds and grasses.
- 27. A method of controlling broad-leaved weeds and grasses, which comprises treating the crop plants which are to be protected against broad-leaved weeds and grasses, or the broad-leaved weeds and grasses themselves, with a composition according to claim 25.
- 28. A pesticidal composition according to claim 25, which is used for controlling insects or representatives of the order Acarina.
- 29. A pesticidal composition according to claim 25, which is used for the selective control of broad-leaved weeds and grasses in crops for useful plants.
- 30. The use of a composition according to claim 26 for controlling insects or representatives of the order Acarina.
- 31. The use of a composition according to claim 26 for the selective control of

broad-leaved weeds and grasses in crops of useful plants.

- 32. A method of controlling insects or representatives of the order Acarina, which comprises applying a composition according to claim 28 to the pests or their habitat.
- 33. A method according to claim 32 for protecting vegetative propagation stock against infestation by insects or representatives of the order Acarina, which comprises treating the propagation stock or the site where the propagation stock is planted out.
- 34. Vegetative propagation stock treated by the method described in claim 33.
- 35. A method for controlling unwanted plant growth, which comprises treating the crop plants which are to be protected against broad-leaved weeds, and/or the broad-leaved weeds and grasses, with a composition according to claim 25.
- 36. A composition according to claim 28, which contains from 0.1 to 99 percent by weight of active substance.
- 37. A method according to claim 32, wherein from 0.001 to 2 kg of active substance are applied per hectare.
- 38. A composition according to claim 29, which contains from 0.1 to 95 percent by weight of active substance of the formula I.
- 39. A method according to claim 35, which comprises applying a compound of the formula I in a quantity of from 0.001 to 2 kg per hectare.
- 40. A selective-herbicidal composition which comprises, in addition to customary inert formulation auxiliaries, a mixture of a herbicidally effective quantity of a compound of the formula I and safener, as active substance.
- 41. A herbicidal composition according to claim 40, wherein the safener is a quinoline derivative, pyrazole derivative or triazole derivative.
- 42. A composition according to claim 41, wherein the safener is either a quinoline derivative of the formula

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$$\begin{array}{c}
X_2 \\
O-CH_2-C-O-R_{15} \\
0
\end{array}$$
(X),

in which

R₁₅ is hydrogen, C₁-C₈alkyl or C₁-C₈alkyl which is substituted by C₁-C₆alkoxy or

C₃-C₆alkenyloxy; and

X₂ is hydrogen or chlorine; or

a 1-phenylazole-3-carboxylic acid derivative of the formula

$$R_{16}$$

$$R_{16}$$

$$R_{17}$$

$$R_{18}$$
(XI),

in which

E is nitrogen or methine;

R₁₆ is -CCl₃ or unsubstituted or halo-substituted phenyl;

R₁₇ and R₁₈ independently of one another are hydrogen or halogen; and

 R_{19} is C_1 - C_4 alkyl.

43. A composition according to claim 40, wherein, in the compounds of the formula I, the

radical R_1 is the group $(R_4)_n = (R_5)_m$; and m is 0.

- 44. A composition according to claim 43, in which R₄ is C₁- or C₂alkyl; and n is 1, 2 or 3.
- 45. A composition according to claim 44, in which R_4 is C_1 or C_2 alkyl in positions 2, 4 and 6 relative to the phenyl linkage site; and n is 3.
- 46. A composition according to claim 40, wherein, in the compounds of the formula I, R₁

is the group $(R_4)_n$; m is 0; R_4 is C_1 - or C_2 alkyl; n is 1, 2 or 3; R_2 and R_3 are

 C_1 - C_6 alkyl; or R_2 and R_3 together form a substituted or unsubstituted alkylene chain -(CH_2)₅-, -(CH_2)₄- or -(CH_2)₃-.

47. A composition according to claim 40, wherein, in the compounds of the formula I, G is the group -CO-A (a); and A is C_1 - C_8 alkyl substituted by nitro, cyano, $Si(C_1$ - C_4 alkyl)₃, amino, C_1 - C_4 alkylamino, $di(C_1$ - C_4 alkyl)amino, unsubstituted or substituted benzyloxy or a group $-\frac{CH-NR_{08}R_{09}}{I}$, or A is C_3 - C_8 cycloalkyl which if desired can contain as $\frac{I}{COOR_{010}}$

heteroatom oxygen, sulfur or nitrogen and is substituted by C_1 - C_4 alkyl, C_1 - C_4 alkoxy, C_1 - C_4 alkylsulfinyl, C_1 - C_4 alkylsulfonyl or substituted or unsubstituted phenyl, or A is C_3 - C_8 cycloalkyloxy which can if desired contain as heteroatom oxygen, sulfur or nitrogen and is unsubstituted or substituted by C_1 - C_4 alkyl, or A is a group - $(CR_8R_9)_p$ - $(Y)_o$ - $(Y)_o$ - $(Z)_r$ - $(CR_8R_9)_p$ - $(Y)_o$ - $(CR_8R_9)_p$ -

48. A composition according to claim 40, wherein, in the compounds of the formula I, R_1 is the group $(R_4)_n$; R_4 is C_1 - or C_2 alkyl; n is 1, 2 or 3; m is 0; R_2 and R_3 are

 C_1 - C_6 alkyl; or R_2 and R_3 together form a substituted or unsubstituted alkylene chain -(CH_2)₅-, -(CH_2)₄- or -(CH_2)₃-; G is the group -CO-A (a); and A is C_1 - C_8 alkyl substituted by nitro, cyano, $Si(C_1$ - C_4 alkyl)₃, amino, C_1 - C_4 alkylamino, $di(C_1$ - C_4 alkyl)amino, unsubstituted or substituted benzyloxy or a group -CH- NR_{08} R₀₉, or A is C_3 - C_8 cycloalkyl $COOR_{010}$

which can if desired contain as heteroatom oxygen, sulfur or nitrogen and is substituted by C_1 - C_4 alkyl, C_1 - C_4 alkoxy, C_1 - C_4 alkylthio, C_1 - C_4 alkylsulfinyl, C_1 - C_4 alkylsulfonyl or substituted or unsubstituted phenyl, or A is C_3 - C_8 cycloalkyloxy which can if desired contain as heteroatom oxygen, sulfur or nitrogen and is unsubstituted or substituted by C_1 - C_4 alkyl, or A is a group - $(CR_8R_9)_p$ - $(Y)_o$ -CO- $(Z)_r$ - R_{10} or - $(CR_8R_9)_p$ - R_{11} .

49. A composition according to claim 42, which comprises as safener a compound of the formula

$$(Xa).$$

50. A composition according to claim 42, which comprises as safener a compound of the formula

51. A composition according to claim 42, which comprises as safener a compound of the formula

- 52. A method for the selective control of broad-leaved weeds and grasses in crops of useful plants, which comprises treating the crops, their seed or their cultivation area, simultaneously or independently of one another, with an effective quantity of a herbicide of the formula I and a herbicide-antagonistically effective quantity of a safener of the formula X or XI according to claim 42.
- 53. A method according to claim 52, which comprises treating stocks of crop plants or cultivation areas for crop plants with from 0.001 to 2 kg/ha of a herbicide of the formula I and a quantity of from 0.001 to 0.5 kg/ha of a safener of the formula X or XI.
- 54. A method according to claim 52, wherein the crop plants are maize and cereals.

- 55. A compound according to claim 1 of the formula I, in which R_1 is 2,4,6-trimethylphenyl, R_2 and R_3 together are -(CH₂)₄- and G is -C(=O)-N(CH₃)CH₂CH₂CN.
- 56. A compound according to claim 1 of the formula I, in which R₁ is 2,4,6-trimethylphenyl, R₂ and R₃-N together are -CH₂CH₂CH(CH₃)CH₂-N and G is -C(=0)-N(CH₃)CH₂CH₂CN.
- 57. A compound according to claim 1 of the formula I, in which R_1 is 2,6-dichlorophenyl, R_2 and R_3 together are -(CH₂)₄- and G is -C(=O)-N(CH₃)CH₂CH₂CN.

INTERNATIONAL SEARCH REPORT



In Application No 95/05152

A. CLAS	SIFICATION OF SUBJECT MATTER C07D231/32		
According	to International Patent Classification (IPC) or to both national c	lassification and IPC	
	S SEARCHED		
Minimum IPC 6	documentation searched (classification system followed by classi CO7D	fication symbols)	
170 0			
Documents	ation searched other than minimum documentation to the extent t	hat such documents are included in the fields	searched
	data base consulted during the international search (name of data	base and, where practical, search terms used)	
C. DOCUM	MENTS CONSIDERED TO BE RELEVANT		
Category *	Citation of document, with indication, where appropriate, of the	e relevant passages	Relevant to claim No.
Y	EP,A,O 508 126 (BAYER AG) 14 Occited in the application see page 44 - page 45; claim 1	tober 1992	1-57
Y	DE,A,44 13 669 (BAYER AG) 12 Ja see page 22 - page 23; claim 1	nuary 1995	1-57
Y	EP,A,O 355 599 (BAYER AG) 28 Fe see page 51 - page 52; claim 1 see page 29; example 144; table		1-57
Furt	ner documents are listed in the continuation of box C.	Patent family members are listed in	n annex.
* Special cat	egories of cited documents :	"T" later document published after the inte	mational filing date
	ent defining the general state of the art which is not cred to be of particular relevance	or priority date and not in conflict wi cited to understand the principle or th invention	th the application but
"E" earlier of filing d	document but published on or after the international late	"X" document of particular relevance; the cannot be considered novel or cannot	be considered to
which i	nt which may throw doubts on priority claim(s) or is cited to establish the publication date of another	involve an inventive step when the do "Y" document of particular relevance; the	cument is taken alone claimed invention
	or other special reason (as specified) intreferring to an oral disclosure, use, exhibition or	cannot be considered to involve an in- document is combined with one or me ments, such combination being obvious	ventive step when the ore other such docu-
P' docume	nt published prior to the international filing date but an the priority date claimed	in the art. '&' document member of the same patent	
Date of the a	actual completion of the international search	Date of mailing of the international sea	arch report
20) March 1996	. :.	
Name and m	alling address of the ISA	Authorized officer	
	European Patent Office, P.B. 5818 Patentiaan 2 NL - 2280 HV Rijswijk Tel. (+ 31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+ 31-70) 340-3016	Fink, D	

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INTERNATIONAL SEARCH REPORT

Information on patent family members

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